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The American Mineralogist

Journal of the Mineralogical Society of America

Vol. 38 JANUARY-FEBRUARY, 1953 Nos. 1 and 2

Dedicated to

WALTER F. HUNT

Scholar and Teacher, and
Editor of the American Mineralogist Since 1922

By His Colleagues and Students at the University of Michigan on the occasion of his becoming Professor Emeritus, September 1952

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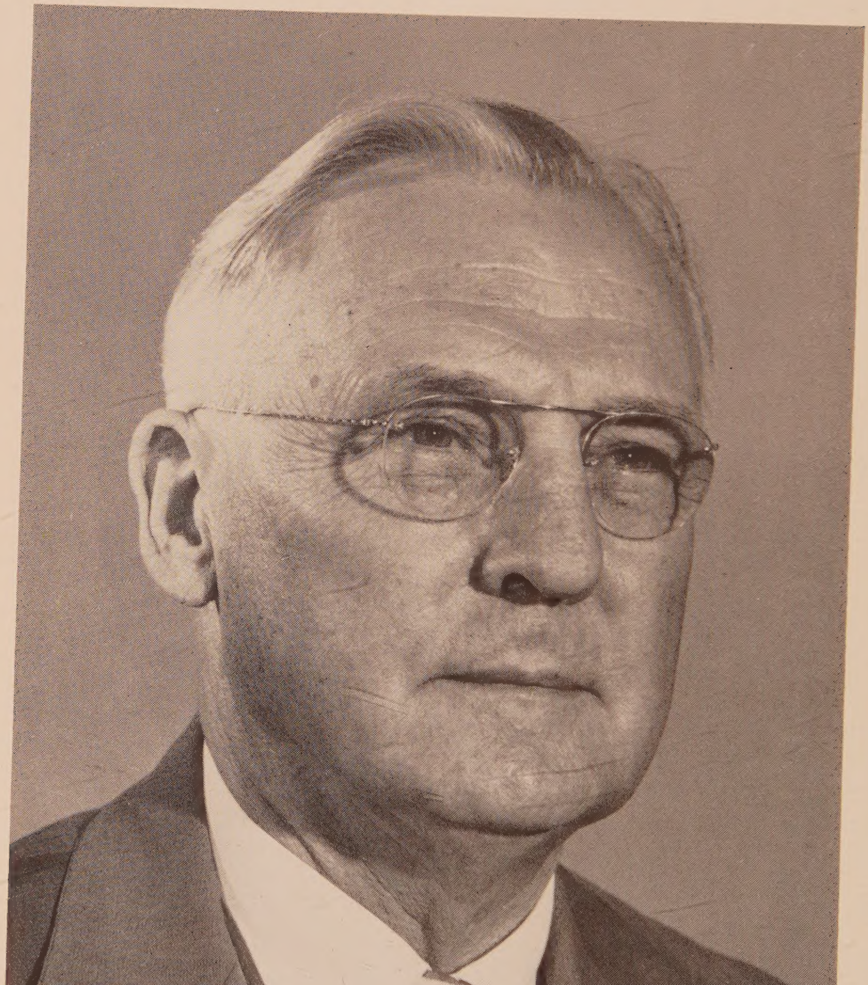

Walter F. Hunt

THE AMERICAN MINERALOGIST

JOURNAL OF THE MINERALOGICAL SOCIETY OF AMERICA

Vol. 38 JANUARY-FEBRUARY 1953 Nos. 1 and 2

WALTER FREDERICK HUNT

EDWARD H. KRAUS

This number is dedicated by his colleagues and students of the Department of Mineralogy to Walter Frederick Hunt, upon his retirement from active duty to the University of Michigan, in deep appreciation of his service as a teacher and investigator, and as Editor of the *American Mineralogist* since 1922.

Professor Hunt was born in Cincinnati, Ohio, September 6, 1882. After attending the public schools in that city, he enrolled in the University of Michigan where as an undergraduate he specialized in chemistry. After receiving the degree of Bachelor of Arts in 1904 he immediately entered upon graduate work and became interested in mineralogy. He was soon appointed to the staff of the department and rose through the various ranks to Professor of Petrology in 1922 and as Chairman of the Department and Director of the Mineralogical Laboratory in 1933. The Degree of Doctor of Philosophy was conferred upon him in 1915. In 1951 he was on retirement furlough and became professor emeritus in September, 1952. Professor Lewis S. Ramsdell has succeeded him as Department chairman.

As a teacher Professor Hunt was extremely painstaking and meticulous. He is an excellent lecturer and never met a class without careful preparation. He always personally supervised his laboratory courses and in all his teaching he demanded good performance by his students. By his colleagues and by those who came under his instruction he is held in very high esteem for his comprehensive knowledge and his sympathetic attitude.

Dr. Hunt early demonstrated a flair for research as is evidenced by a very creditable list of published papers dealing with various phases of crystallography and mineralogy. For many years he prepared hundreds of abstracts of mineralogical, chemical, and geological papers which were published in *Chemical Abstracts* in this country and in journals abroad. He also collaborated in publishing two texts,—*Tables for the*

Determination of Minerals (with E. H. Kraus) and *Mineralogy, An Introduction to the Study of Minerals and Crystals* (with E. H. Kraus and L. S. Ramsdell).

In 1922 Professor Hunt became Editor of the *American Mineralogist* which had been taken over as the *Journal* of the Mineralogical Society of America when the society was organized in 1919. The *American Mineralogist* was first issued in 1916 as an independent journal under the editorship of Wallace Goold Levison. He was succeeded by Edgar T. Wherry in 1919 who continued in office for three years. That there was need for the *American Mineralogist* is shown by its annual growth in those early years. The first volume (1916) was a slender one and consisted of xvi+102 pages. As its availability for publishing mineralogical and crystallographic papers became known there was a material increase in the size of the annual volumes. Thus, during the first year of Professor Hunt's editorship, Volume 7 (1922) consisted of xii+214 pages.

Since the organization of the Mineralogical Society in December, 1919, interest in mineralogy and related fields in the United States and Canada has increased greatly. This is evidenced by the present large membership in the Society, which totals 1134 (5 correspondents, 315 fellows, 814 members) and 799 subscribers, as compared with a total of 181 (55 fellows and 126 members) and 139 subscribers at the close of the first year (1920). During this period of over three decades there has been a corresponding growth in the other fields of the earth sciences for several new societies were organized all of which have had remarkable growth.

The *American Mineralogist* has been a powerful influence in stimulating the publication of the results of researches, notes and news, book reviews, and the proceedings of societies. Mention must also be made of several important special numbers, such as the *Symposium on Quartz Oscillator-Plates*, those honoring Professors Charles Palache and Esper S. Larsen, Jr., and the ones devoted to Canadian mineralogy. Volume 37 (1952) which has just been published is very large and consists of vi+1088 pages.

The task of editing a highly specialized scientific journal is a difficult one. Contributions are received from scientists from all over the world who recognize the importance of publication in the *American Mineralogist*, which maintains very high standards and has a wide circulation. Editor Hunt has long demonstrated that he has an excellent over-all knowledge of the fields served by the journal, as is clearly revealed by the high character of the articles accepted for publication.

It is obvious that an editor cannot approve of all articles which are submitted, for some do not come within the fields to which the journal is devoted, and others do not meet the standards set for publication. In these matters an editor must be extremely considerate, fair, and diplomatic. Then too, editors cannot be "thin skinned" for it is absolutely impossible to satisfy everyone. It has long been shown by the prestige of the *American Mineralogist* and its appeal to the scientists the world over, that Professor Hunt is a very successful and efficient editor.

Only those who have been and now are closely associated with him know how heavily the importance of maintaining high standards and the widening of the influence of the journal weigh upon him. Very few realize the long hours he personally devotes to the task of preparing manuscripts for the printer and to subsequently correcting the proof. This is all done primarily for the good of our science and not for pecuniary gain. The present cost of editing our journal is very small, indeed much smaller than for other journals of like character and size.

Professor Hunt was a member of the small group of mineralogists who were active in organizing the Society and hence is a charter fellow. Moreover, he has been active in the administrative affairs of the organization longer than anyone else for he has been a member of the Council continuously since he became Editor in 1922, thirty-one years ago.

I am confident that the readers of the *American Mineralogist* agree with the staff and students of the Department of Mineralogy of the University of Michigan that this special number is a fitting and well-merited expression of appreciation of the long service that Walter F. Hunt has rendered to the advancement of mineralogical and related sciences. It is our sincere hope that many more fruitful years may be in store for him.

Professor Hunt prepared the manuscripts for the printer and corrected proof in the usual manner for this issue. With the cooperation of the publisher, the assistant editor then made certain changes and additions, and Professor Hunt was not aware that this issue would be dedicated to him.

L. S. R.

HUNTITE, $Mg_3Ca(CO_3)_4$, A NEW MINERAL*

GEORGE T. FAUST, *U. S. Geological Survey, Washington, D. C.*

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ABSTRACT

Huntite, $Mg_3Ca(CO_3)_4$, a new mineral from Currant Creek, Nevada, is named in honor of Professor Walter F. Hunt. This mineral was deposited by cool waters in cavities and vugs in rocks composed of magnesite, dolomite, and deweylite. Electron micrographs of huntite show that it is very fine-grained (1 micron and less). The *x*-ray powder diffraction data permit it to be differentiated from the other carbonates of magnesium and calcium. The principal lines in the powder pattern are: 2.838,VVS; 1.986,S; 1.769,S; 2.580,M; 2.444,M; 2.388,M; 2.204,M; 1.590,M; 1.488,M; 1.010,M. All values are expressed in Å.

The unit cell data are as follows: orthorhombic, primitive lattice; $a_0=5.66$ Å, $b_0=9.02$ Å, $c_0=8.51$ Å; 2 formulas per cell.

INTRODUCTION

The mineral described in this paper was first found in 1943 while the author was making a study of the distribution of dolomite in magnesite specimens from Currant Creek, Nevada, by means of the flame test for calcium. The physical appearance of this mineral suggested that it was magnesite, but the flame test showed calcium and accordingly the mineral was studied in more detail. The mineralogy and petrology of the

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* Publication authorized by the Director, U. S. Geological Survey.

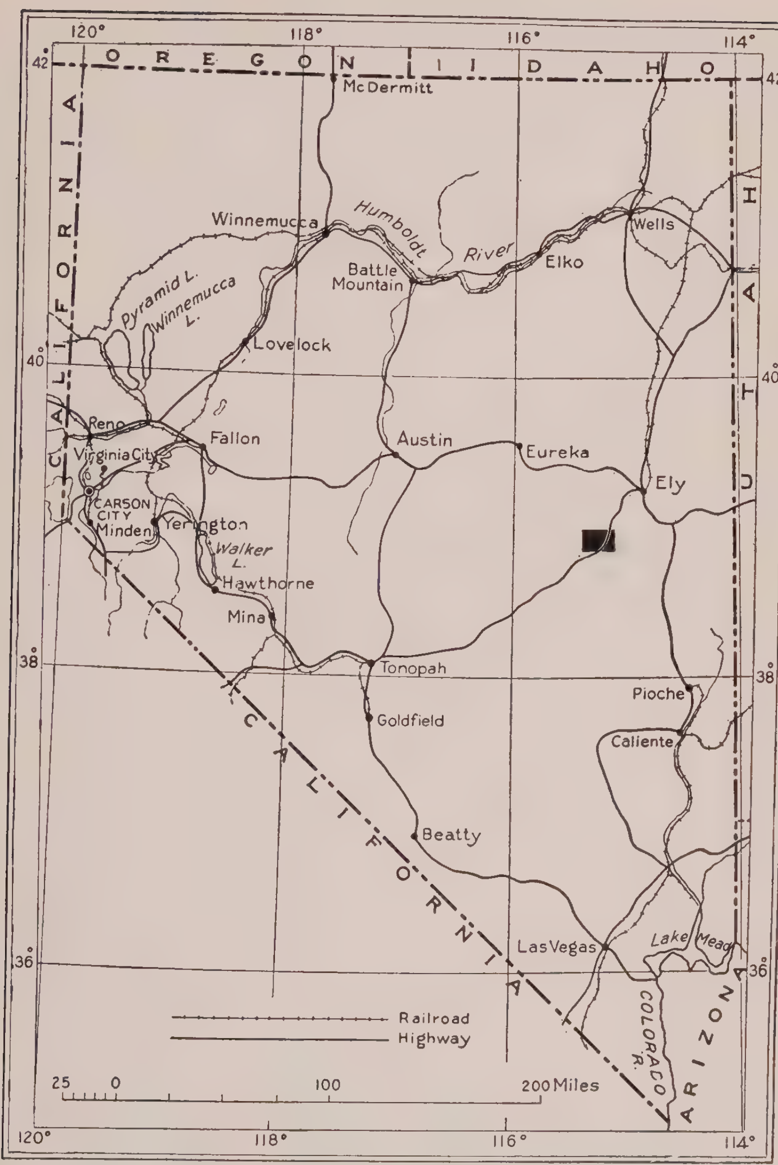

FIG. 1. Index-map of Nevada showing the location of the Currant Creek magnesite deposits.

deposits at Currant Creek were described by Faust and Callaghan (1948), and the material was tentatively considered to be a solid solution of magnesite and dolomite. A brief description of the mineral was included in that paper and its position in the sequence of the minerals was established. Further studies on this mineral showed that it is a new species. In September 1950, the mineral was identified in a sample submitted by Dr. T. S. Lovering from the north end of the Longyear Exploration Drift in the 1800 level of the Chief Consolidated Mine, Eureka, Utah.

It is possible that huntite has been observed before and considered to be impure magnesite. Evidence for such belief is given by Ford (1917, p. 221): "There is one analysis of magnesite quoted with some 23 per cent of calcium carbonate."

Locality

The Currant Creek magnesite deposits are located in White Pine and Nye Counties, Nevada, about 29 miles southwest of Ely, Nevada (Fig. 1). This district contains seven prospects where development work has been done (Vitaliano, 1951). The prospects are the Chester, Windous, and Rigsby deposits in the eastern part of the area, in White Pine County, the Nevada Magnesite (sometimes referred to as the Rex-Pine Lode), Snowball, and White Knolls deposits in the western part of the area, in White Pine County, and the Ala-Mar deposit in Nye County. Huntite has been found at the Ala-Mar, Nevada Magnesite, and Snowball deposits.

Occurrence and Paragenesis

(1) *Geological environment*

The Currant Creek magnesite deposits are located in the Horse Range, a group of relatively low hills of Tertiary volcanic rocks paralleling the northwest-trending White Pine Range on its east side. The layers of volcanic rock dip gently eastward away from the Paleozoic rocks of the White Pine Range, but are broken by many faults. The Paleozoic rocks are directly overlain by a series of flows consisting chiefly of dacite; in the western part of the area, near the Ala-Mar deposit, the dacite is overlain by volcanic breccia, tuffaceous in places, which in turn is overlain by basaltic andesite having a vesicular and basaltic appearance at the top. The dacite, volcanic breccia, and basaltic andesite are grouped together as the lower volcanics. The host rock of the magnesite and associated minerals is a bedded tuff formation, the Currant tuff, a few feet to more than 400 feet thick, which overlies the lower volcanics. The Currant tuff is overlain by latite which is exposed over a large part of the district, but east of the Ala-Mar mine it is overlain by latite crystal tuff. The latite and latite crystal tuff are grouped together as the upper volcanics. The association of magnesite-bearing areas with faults suggests that the faults provided the channel ways for the ascent of the magnesium-bearing solutions.

The Currant tuff was named by Faust and Callaghan (1948) for the Currant Creek area, Nevada, where it is well exposed; their paper contains full descriptions of the mineralogy and petrography of these tuffs, and the reader is referred to it for additional details. The minerals of the tuffs include hypersthene, augite, hornblende, biotite, plagioclase of various compositions, sanidine, quartz, apatite, magnetite, and calcite, all intimately mixed with glass shards and irregular glass fragments. The reworked members of the Currant tuff are calcareous and grade into tuffaceous limestones. Subsequent to deposition some of the tuff beds were altered by cool meteoric waters. The volcanic glass was in part changed to montmorillonite and there was some replacement by calcite.

The magnesite deposits were formed by hydrothermal solutions rich in magnesium and carbon dioxide which ascended along channelways provided by the fault zones. These solutions entered the tuff beds and preferentially attacked the calcite-bearing tuffs. Where calcite was present, the replacing solutions produced dolomite by reaction until no more calcite was in actual contact with the solution. Thereafter, the hydrothermal solutions were free to replace the tuffs with magnesite and deweylite. These rocks, formed by the action of the hydrothermal solutions on the tuffs, are designated as the replaced tuffs in this paper.

(2) *Order of succession of the minerals in the magnesite deposit*

The paragenetic sequences of the minerals in several specimens from the magnesite deposits are shown in Fig. 2. These specimens are fully described by Faust and Callaghan (1948). The composite paragenetic sequence is shown in Fig. 3. The early stage represents normal replacement of the tuffs by calcite and montmorillonite through the agency of ground waters. The hydrothermal stage represents, at first, the reaction of the magnesium-rich solutions on the calcite of the tuffs to form dolomite. When the solutions became impoverished in calcium, magnesite and, later, deweylite were formed. Thereafter, the solutions spread into the overlying calcareous tuffs, converted the calcite to dolomite and deposited deweylite. At the close of the hydrothermal stage, calcite and silica minerals (quartz, chalcedony, opal) were deposited along with some dolomite and deweylite in the upper tuff beds; finally the silica minerals and calcite were the sole phases forming. The solutions gradually cooled to the temperature of the ground waters and calcite continued to form. Some of the replaced rock has a very vuggy texture (Fig. 4). The vugs are commonly almond-shaped and range in size from 10 cm. down to microscopic dimensions. The matrix of the replaced rock is fine-grained, dense and in some specimens porcelaneous. During the late meteoric stage the vugs in the replaced rocks were lined with botryoidal calcite which

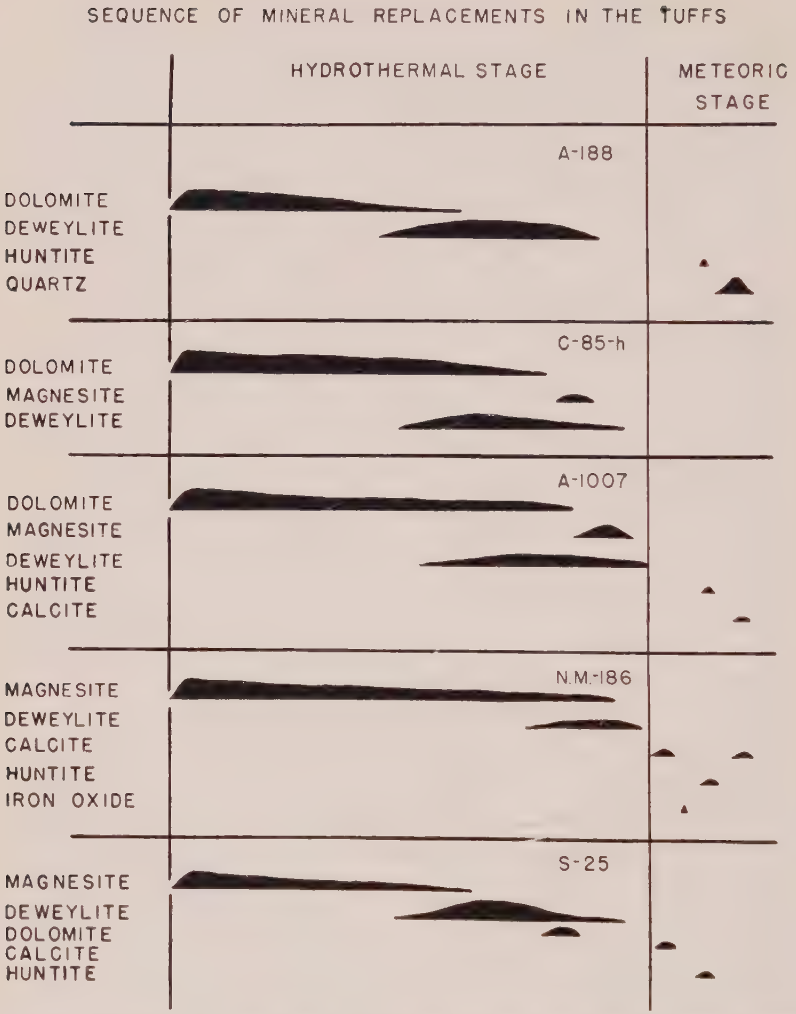

FIG. 2. Sequence of replacement for individual specimens which represent various stages in the formation of the magnesite deposits. The vertical superposition in the figure of the various sequences does not imply that replacement began and ended at the same time for all the specimens represented.

in turn was later stained yellow to brown by a thin film of iron oxides. The last mineral to fill these cavities is huntite, which occurs as a white fine-grained powdery mass; because of its white color, the huntite contrasts conspicuously with the matrix (Fig. 5). Some samples are encrusted by calcite.

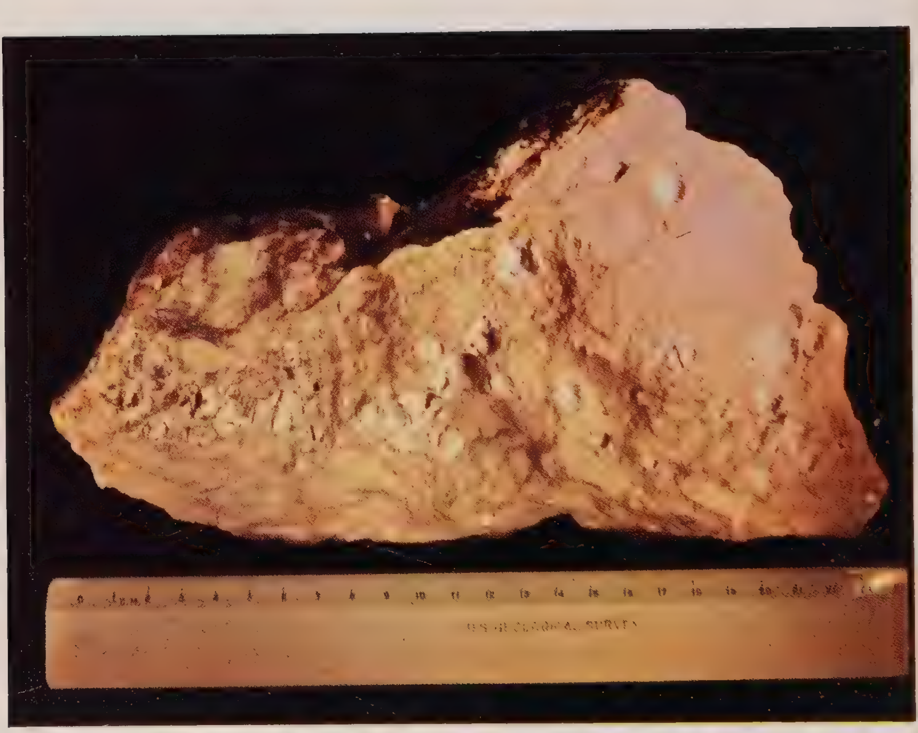

Fig. 5. Replaced tuff N.M.-186. Huntite filling vugs in buff matrix. The matrix is magnesite with some deweylite.

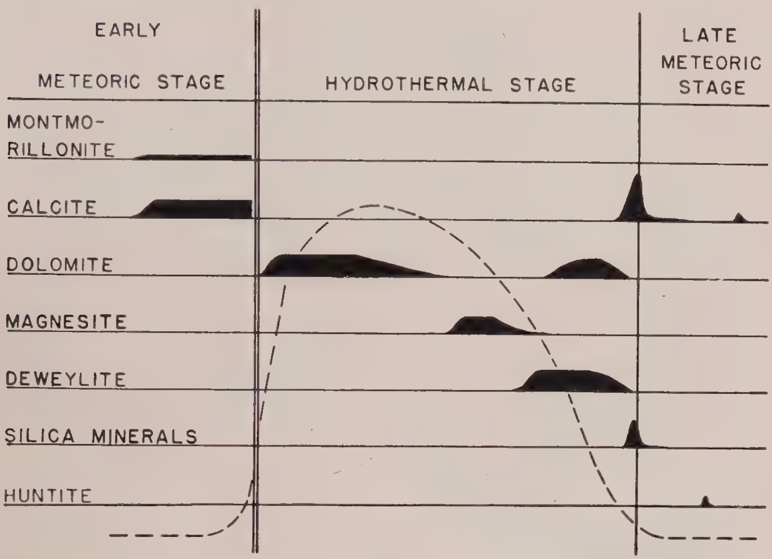

FIG. 3. Composite chart showing the paragenesis of the minerals of the magnesite deposits.

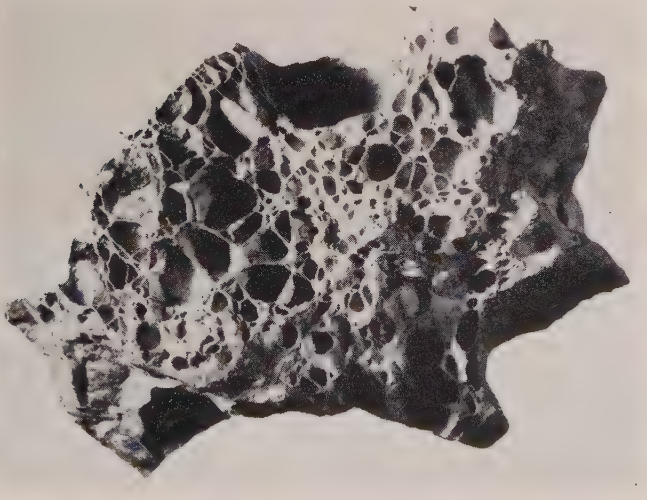

FIG. 4. Vuggy texture developed in microcrystalline magnesite rock R-99, Rigsby deposit, Currant Creek, Nevada. Specimen two-thirds natural size.

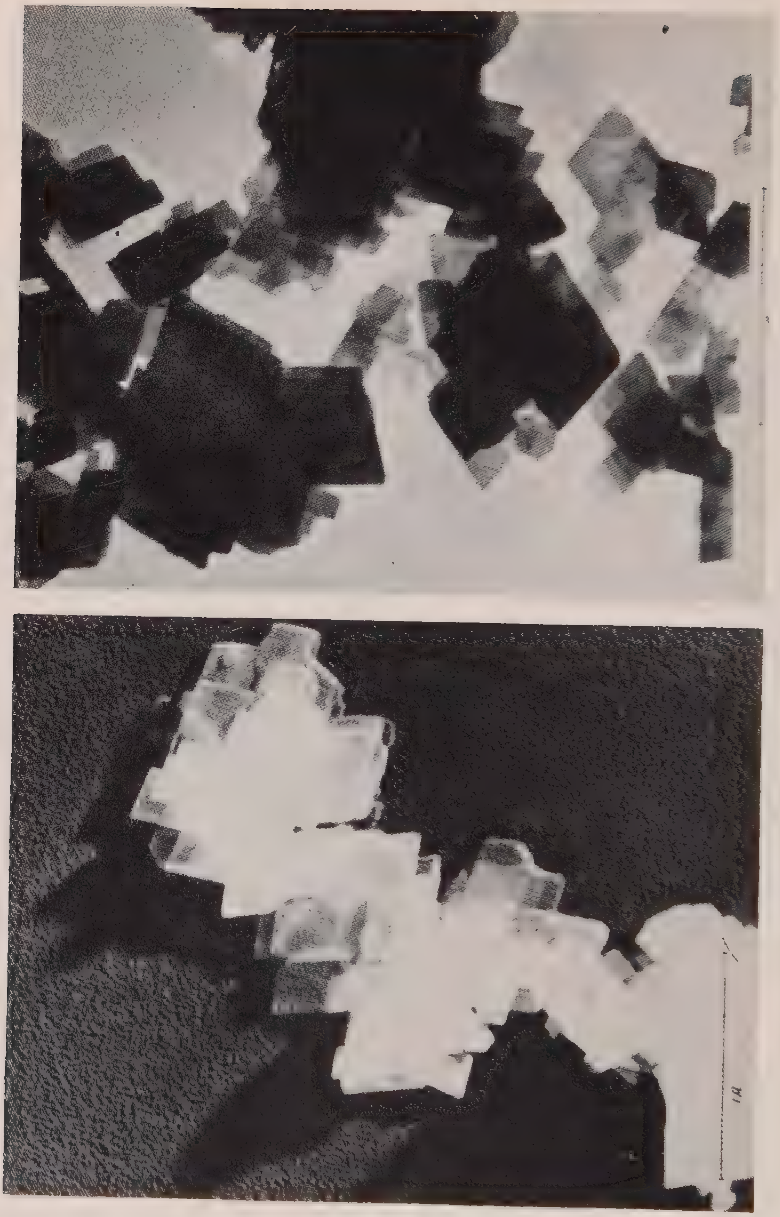

FIG. 6. Electron micrographs of huntite. The upper photograph demonstrates the homogeneity of the sample (magnification approximately 32,000×). The lower photograph shows the crystals of huntite shadowed with chromium at an angle of 9°35′ (magnification approximately 30,000×). Electron micrographs prepared by E. J. Dwornik.

Huntite also occurs as a white powdery mass at the base of the replaced tuffs where the replaced tuff is in contact with vitric tuff.

(3) *Origin of huntite*

The paragenetic sequences for various specimens suggest that huntite was formed during the late meteoric stage. It is definitely younger than the calcite and iron oxides lining the vugs but it is older than some of the calcite. It is believed that huntite was precipitated as a very fine powder from cool ground waters which gathered their magnesium locally in traversing the magnesite deposits. It is believed that huntite is a metastable phase and experimental work is in progress to determine its range of stability.

CRYSTALLOGRAPHIC STUDIES

(1) *Morphology*

The crystals of huntite were judged to be exceedingly small, both from the microscopic examination of the powder and from the similarity of its behavior to that of a fine-grained chemical precipitate which has been redispersed in water. Accordingly, the powder was examined by electron microscopy, with most gratifying results. The electron micrographs of huntite prepared by E. J. Dwornik are shown in Fig. 6. The size of the largest crystals appears to be about one micron, measured in the direction of the longest diagonal. These micrographs suggest that huntite has a platy habit.

(2) *X-ray powder diffraction data*

Powdered huntite was mixed with "Duco" cement as a binding medium and formed into a spindle. A powder pattern was obtained using filtered copper radiation. The data for the interplanar spacings, *d*-spacings, were corrected according to the method of Straumanis and Ievins (1940); the results are given in Table 1, together with the data for dolomite, magnesite, calcite, aragonite, and vaterite. A comparison of these data shows clearly the difference between huntite and the other carbonates.

(3) *X-ray crystallographic study*

Although the visual comparison of the powder photograph of huntite with those of dolomite, magnesite, and a mixture of dolomite and magnesite (specimen W-300; estimated composition magnesite 93%, dolomite 7%), see figure 7, removes any doubt of its uniqueness, it is very desirable to index the lines in the *x*-ray diffraction pattern.

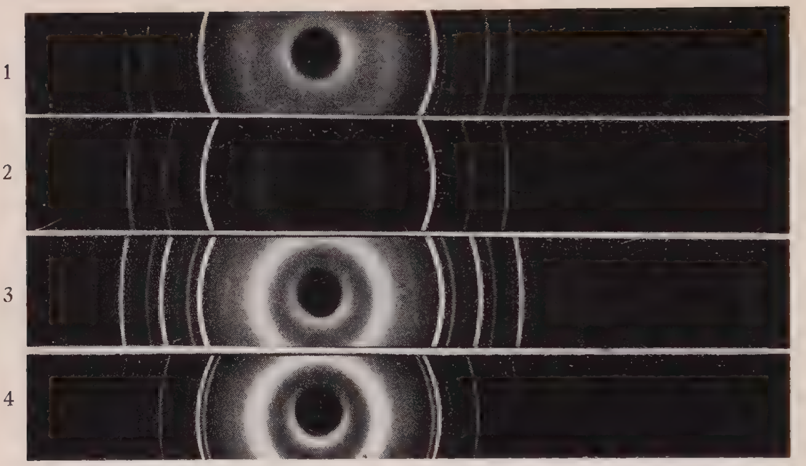

FIG. 7. 1. Huntite. 2. Dolomite. 3. Magnesite. 4. Dolomite 7%, magnesite 93%.

Dr. W. F. Bradley of the Illinois Geological Survey suggested that an attempt be made to index the lines in the powder pattern on the assumption of a mixture consisting of a solid solution of magnesite in dolomite and of a solid solution of dolomite in magnesite. The results of lengthy calculations failed to provide any good agreement between the observed and calculated values of the *d*-spacings for two hexagonal unit cells. The application of the requirements of a rhombohedral unit cell, that the algebraic sum of the hexagonal indices satisfy the relations: (1) $-h+k+l=3n$, or (2) $h-k+l=3n$, eliminated a very large number of the possible agreements. It should be noted that the *intensity* of the 2.838 (line) stands in marked contrast to all other lines in the pattern of huntite and this relationship serves to differentiate huntite from the other carbonates.

Dr. Howard T. Evans examined a series of electron micrographs of huntite and suggested (1) that the huntite crystals may be orthorhombic with a platy habit, and (2) that the 2.838 Å line may correspond to an $00l$ reflection.

Accordingly, an attempt was made to index the *d*-spacings on an orthorhombic cell. By indexing the strongest *d*-spacing as 003 and using the charts for orthorhombic crystals prepared by J. O. Wilhelm (1927), two possible solutions, yielding two different axial ratios, were found. A possible morphological "axial ratio" was derived from the measurements of the diagonals of a number of the "diamond-shaped" plates in the electron micrographs. The two axial ratios obtained from the charts of Wilhelm were qualitatively related by simple multiples to the possible morphological "axial ratio" derived from the electron micrographs. Next,

TABLE 1. *X*-RAY POWDER DATA FOR HUNTITE AND OTHER ANHYDROUS CARBONATES OF CALCIUM AND MAGNESIUM

Huntite (*A*-1007-*a*), Ala-Mar deposit, Current Creek, Nev.		Dolomite, data from Mehmel (1939)		Magnesite (W-180), Windous deposit, Currant Creek, Nev.		Calcite, data from Mehmel (1939)		Aragonite, data from Mehmel (1939)		Vaterite, data from Brooks, Clark, and Thurston (1950)	
d(Å)	*I*	*d*(kX)	*I*	*d*(Å)	*I*	*d*(kX)	*I*	*d*(kX)	*I*	*d*(kX)	*I*
5.69	VVVVW										
4.81	VVVVW					4.28	W				
4.16	VVVVW										
		3.81	W			3.86	MW				
3.67	VVVVW										
				3.56	VF					3.59*	
						3.40	M				
								3.39	S		
										3.285	
								3.23	M		
3.15	VVVVW	3.18	W								
				3.04	W	3.06	VS				
2.905	W										
2.838	VVS	2.85	VS								
				2.738	VS					2.719	
		2.65	MS					2.68	MS		
2.580	M										
		2.53	W			2.52	M				
				2.508	S						
								2.47	M		
2.444	M										
2.388	M	2.39	MW								
				2.318	M	2.31	MS	2.34	S		
2.298	W										
2.204	M										
		2.18	MS					2.18	W		
		2.00	M	2.099	M	2.11	MS	2.09	MW	2.060	
1.986	S†										
								1.958	S		
				1.939	S						
						1.929	S				
1.900	VVW										
				1.884	VF	1.887	S				
								1.860	MS		
1.840	VVW										
		1.837	W								
1.803	VVW					1.807	W	1.800	M		
		1.780	S								
1.769	S			1.771	WM						
								1.720	S		
1.708	VVW			1.701	VS						
						1.689	W				
						1.620	M			1.644	

Huntite (*A*-1007-*a*), Ala-Mar deposit, Current Creek, Nev.	Dolomite, data from Mehmel (1939)	Magnesite (W-180), Windous deposit, Currant Creek, Nev.	Calcite, data from Mehmel (1939)	Aragonite, data from Mehmel (1939)	Vaterite, data from Brooks, Clark, and Thurston (1950)
1.590 M					
	1.560 W			1.550 W	
1.529 VW	1.532 M		1.535 M		
		1.510 M			
1.488 M		1.487 M	1.488 W		
				1.460 W	
1.453 W	1.456 MW		1.451 MS		
1.423 VVW	1.427 W				
1.403 VVW		1.406 M		1.400 W	
1.378 VVW	1.382 M	1.372 VW			
1.360 VVW		1.355 M		1.350 W	
		1.336 S			
1.322 VW	1.329 M				
1.311 W			1.307 W		
1.284 VVVW	1.291 W				
1.263 VVVW	1.265 MW				
1.245 W		1.251 WM		1.250 W	
	1.230 M	1.238 VVW		1.230 M	
1.222 VW					
	1.195 W	1.201 VW		1.188 W	
		1.180 W			
1.179 VW			1.181 W		
				1.180 W	
1.167 VVVW	1.162 M				
		1.159 VF	1.159 W		
				1.168 W	
1.148 VVVW			1.150 W		
	1.139 W				
1.131 VVW		1.128 VF			
	1.119 MW			1.120 W	
	1.105 M	1.101 VF		1.100 W	
1.081 VVVW	1.093 MW	1.067 M			
		1.051 W	1.050 M	1.030 MW	
1.010 M	1.002 MW	1.014 VF	1.014 MW	1.000 MW	
0.9771 W					
		0.9683 M			
		0.9563 M			
0.9538 VW					
0.9478 VVVW					
		0.9449 VVF			
0.9297 VVW					
0.9176 VVW		0.9184 VW			
		0.9128 S			
0.8971 VVW		0.9113 W			
		0.8930 VVF			

* Strong reflections only; other reflections not certain.
† Doublet.

the volume of cells containing one, and two molecules of $Mg_3Ca(CO_3)_4$ respectively, were calculated from the measured specific gravity.

With these data, the value of the axial ratio was refined until a good correspondence between the observed and the calculated *d*-spacings was obtained. The results are given in Table 2 which shows that all of the lines can be indexed on the basis of an orthorhombic cell. Donald H. Richter and T. Woodward assisted the author in the numerical computation of the data.

The dimensions of the unit cell obtained from the calculations are: $a_0 = 5.663$ Å, $b_0 = 9.020$ Å, $c_0 = 8.514$ Å. The number of molecules per unit cell, Z, equals 2. The morphological axial ratio derived from the *x*-ray data are $a:b:c = 0.6278:1:0.9439$. A survey of the 43 reflections present in the pattern shows that the lattice of huntite is primitive (*P*). The data available are, however, not sufficient to determine the space group.

Physical Properties

Huntite from the Currant Creek district, Nevada, occurs as compact, chalklike masses which have an even to subconchoidal fracture. It is brittle when cut with a knife and is soft enough to leave a mark on a dark surface. The compact masses are easily cut by the fingernail. It is porous and a fragment dropped into water crumbles, emitting crackling noises as it breaks into smaller fragments. This crackling is accompanied by a hissing sound produced by the displacement of air bubbles from the porous mass. Huntite has a smooth feel and adheres to the tongue.

The specific gravity was determined by J. J. Fahey, using a pycnometer of the type referred to as a "milk test bottle," and a sample weighing 3.8 grams. The specific gravity was found to be 2.696 at 4° C.

Optical Properties

Huntite is white; in thin section it is colorless. Its luster is nonmetallic and dull, and its streak is white. It is nonfluorescent when examined under the radiation from a "Mineralite."

No determinations of the optical properties of huntite were possible by the usual immersion techniques, owing to the very small size of the crystals. Examination in thin section shows, as would be expected, that the birefringence is very strong. When examined in immersion liquids of index of refraction 1.615 and 1.620, some of the aggregates show color fringes.

Differential Thermal Analysis

The equipment and technique used in this laboratory in differential thermal analysis studies, together with references to the literature, are

TABLE 2. COMPARISON OF OBSERVED AND CALCULATED *d*-SPACINGS FOR HUNTITE INDEXED ON AN ORTHORHOMBIC CELL

I	*d* (observed)	*d* (calculated)	*hkl*
VVVVW	5.69	5.66	100
VVVVW	4.81	4.82	110
VVVVW	4.16	4.19	111
VVVVW	3.67	3.54	120
VVVVW	3.15	3.20	112
W	2.91	3.00	030
VVS	2.838	2.840	003
M	2.580	2.574	211
M	2.444	2.447	113
M	2.388	2.397	220
W	2.298	2.306	221
M	2.204	2.208	123
S	1.986	1.992	104, 042, 231*
VVW	1.900	1.887	300
VVW	1.840	1.841	301
VVW	1.803	1.802	050
S	1.769	1.765	043
VVW	1.708	1.707	321
M	1.590	1.591	224
VW	1.529	1.533	125
M	1.488	1.484	323
W	1.453	1.453	160
VVW	1.423	1.425	341
VVW	1.403	1.402	016
VVW	1.378	1.376	106, 054
VVW	1.360	1.361	116
VW	1.322	1.321	145
W	1.311	1.311	261
VVVW	1.284	1.283	036
VVVW	1.263	1.264	305
W	1.245	1.246	352
VW	1.222	1.221	226
VW	1.179	1.179	117, 404
VVVW	1.167	1.167	433
VVVW	1.148	1.148	173
VVW	1.131	1.130	272
VVVW	1.081	1.082	174, 415
M	1.010	1.011	175
W	0.9771	0.978	426
VW	0.9538	0.953	076, 543
VVVW	0.9478	0.948	534, 148
VVW	0.9297	0.930	472
VVW	0.9176	0.918	417, 621
VVW	0.8971	0.897	209, 293

* This line is a doublet.

given by Faust (1948 and 1950). The samples were heated at the rate of approximately 12° C. per minute. The records were obtained as photographs.

(1) *Description of the specimens studied*

The descriptions of the calcite, magnesite, and dolomite standards have been given in an earlier volume of this Journal (Faust, 1949, p. 793). Huntite and other minerals used in the differential thermal analysis examinations are described below.

Huntite. Locality: Ala-Mar deposit, sec. 34, T. 12 N., R. 59 E., Nye County, Nevada. Specimen *A*-1007*a*, compact mass. A portion of the tube sample used in the analysis reported in Table 3 (see under Chemical properties) was used for the differential thermal analysis examination.

Magnesite, dolomite, and deweylite. Locality: Windous deposit, White Pine County, Nevada. Specimen *W*-199, snow-white porous masses, full of cavities. A chemical analysis of this specimen by J. G. Fairchild is given by Faust and Callaghan (1948). Mode obtained from chemical and petrographic data shows: magnesite 64 per cent, dolomite 22 per cent, and deweylite 14 per cent. The ratio of magnesite to dolomite is 75:25.

Magnesite and dolomite. Locality: North Windous tunnel, Windous deposit, White Pine County, Nevada. Specimen *W*-300, white fluffy powder, occurring as a filling in a vug. Euhedral crystals of dolomite are found in the powder. Composition estimated by thermal analysis: magnesite 93 per cent, dolomite 7 per cent.

Mixture of huntite with dolomite and deweylite. Locality: All the material is from the Ala-Mar deposit, Nye County, Nevada. This is an artificial mixture of specimens *A*-1008 and *A*-1007*a* and consists of huntite, dolomite, and deweylite.

(2) *Comparison of differential thermal analysis curves*

The differential thermal analysis curves of huntite, calcite, dolomite, magnesite, and natural and artificial mixtures of these minerals are given for purposes of comparison in Figs. 8 and 9. The curve for huntite (*D*) differs from that of dolomite (*C*) in one very important characteristic, namely, the ratio of the areas under the breaks in the curve. The ratio of the area under the breaks at 792° C. and 926° C. for dolomite from Millville, West Virginia, is 296:510, whereas the ratio of the area under the breaks at 644° C. and 901° C. for huntite from the Ala-Mar deposit is 427:228.

Curve *E* represents the character of the dissociation of a natural mixture, sample W-199 containing 64 per cent magnesite, 22 per cent dolomite, and 14 per cent deweylite. The ratio of magnesite to dolomite in this mixture is 75:25. The clear-cut separation of the break in the curve representing the dissociation of magnesite at 659° C. from the two breaks in the curve arising from the dissociation of dolomite at 806° C. and 885° C. is apparent. The break at 826° C. is due to recrystallization.

Curve *F* is the differential thermal analysis curve for a natural mix-

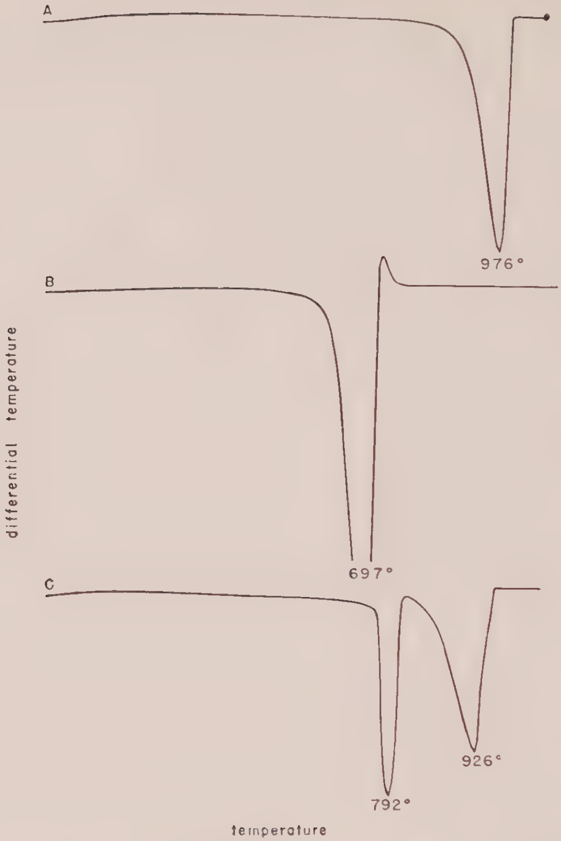

FIG. 8. Differential thermal analysis curves. *A*, calcite no. 13, Joplin, Missouri, record *C*-89, resistance in series with the galvanometer 999.9 ohms. *B*, Magnesite no. 1, Windous deposit, White Pine County, Nevada, record *C*-79, resistance in series with the galvanometer 999.9 ohms. *C*, Dolomite, Millville, West Virginia, Record *F*-3, resistance in series with the galvanometer 900 ohms.

FIG. 9. Differential thermal analysis curves. *D*, Huntite, specimen no. *A*-1007*a*, Ala-Mar deposit, Nye County, Nevada, record *C*-86, resistance in series with the galvanometer 999.9 ohms. *E*. Magnesite and dolomite, with a small amount of deweylite, specimen no. *W*-199, Windous deposit, White Pine County, Nevada, record *C*-151, resistance in series with the galvanometer 600 ohms. *F*, Magnesite 93 per cent, dolomite 7 per cent, specimen *W*-300, vug filling in the North Windous Tunnel, Windous deposit, White Pine County, Nevada, record *C*-152, resistance in series with the galvanometer 600 ohms. *G*, Artificial mixture of 50 per cent huntite (specimen *A*-1007*a*) and 50 per cent of specimen *A*-1008 consisting of dolomite with a small amount of deweylite, both specimens from the Ala-Mar deposit, Nye County, Nevada, record *C*-149, resistance in series with the galvanometer 600 ohms.

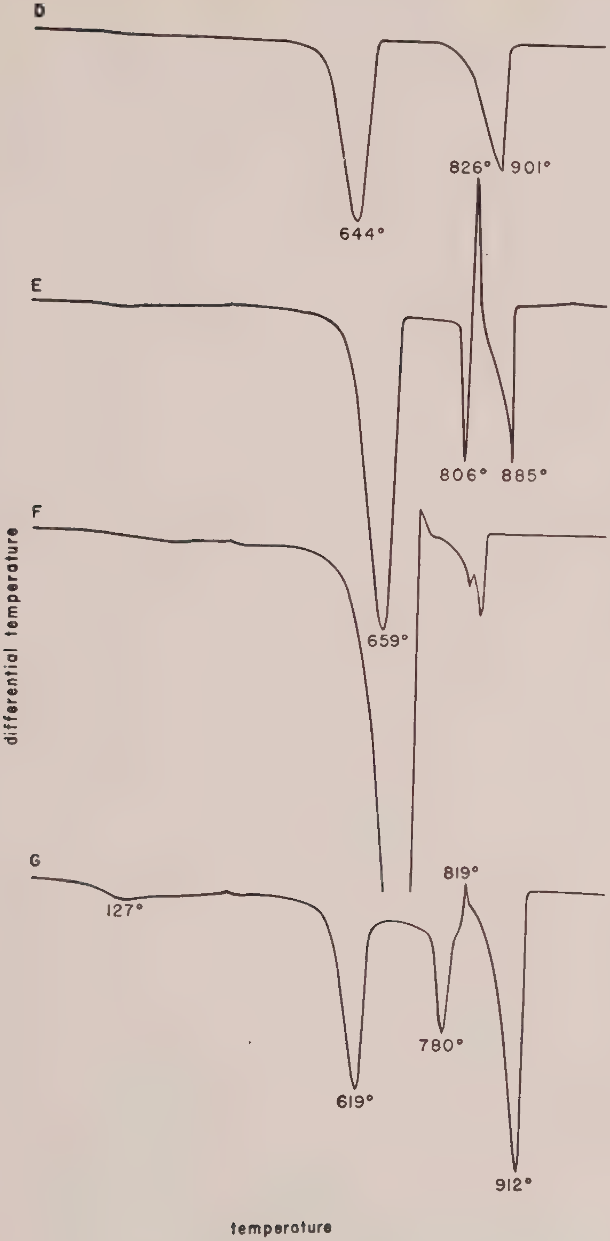
D
826°
901°
644°
E
806°
885°
F
659°
differential temperature
G
127°
819°
780°
619°
912°
temperature

FIG. 9

TABLE 3. MEASUREMENT OF AREA UNDER THE DIFFERENTIAL THERMAL CURVES

Sample	Area of break representing the dissociation of magnesite	Area of break representing the dissociation of $MgCO_3$	Area of break representing the dissociation of $CaCO_3$	Ratio of $MgCO_3$ /$CaCO_3$ in sample	Measured ratio of total area due to $MgCO_3$ /area due to $CaCO_3$	Calculated ratio of total area due to $MgCO_3$ /area due to $CaCO_3$
Dolomite, Curve *C*		296 units	510 units	1:1	0.584	Assumed as 0.60
Huntite, Curve *D*		427	228	3:1	1.87	1.80
Natural mixture of magnesite, dolomite, and deweylite, specimen *W*-199, Curve *E*	753	90*	180	7:1	4.66	4.20

* This area is too large, for it includes the area contributed by the decomposition of deweylite.

ture, sample *W*-300, containing approximately 93 per cent magnesite and 7 per cent dolomite. Here again the breaks in the curve arising from the dissociation of these two minerals is clearly resolved. Curves *E* and *F* should be compared with curve *D* (for huntite).

In order to determine the character of the dissociation of a mixture of dolomite and huntite, a mixture was prepared and subjected to differential thermal analysis. The material mixed with the huntite consists chiefly of dolomite with a small amount of deweylite. This material is similar to the matrix of huntite. The resultant curve *G* is shown in Fig. 9. This curve has three large, distinct, and well-separated breaks. The break at 619° C. represents the dissociation of the $MgCO_3$ in the structure of huntite; the break at 780° C. represents the dissociation of the $MgCO_3$ in the structure of dolomite and, in addition, the dissociation of the small amount of deweylite in the sample; this break is followed by a small exothermic effect at 819° C. due to recrystallization of the constituents in the dissociated deweylite; at 912° C. the break appears for the dissociation of the $CaCO_3$ (derived from the huntite and dolomite).

These thermal analysis curves show that a mixture of dolomite and magnesite can be differentiated from huntite.

(3) *Measurements of areas under the thermal curves*

Areal measurements were made with a simple grid. This grid, approximately 18.5×23.5 cm., is the negative obtained by photographing a piece of high-quality coordinate paper. The areas mentioned in this paper are all given in terms of the smallest unit square. Since we are interested in ratios only, it was not necessary to convert these measurements to square centimeters. The data are presented in Table 3. From the areal measurements was obtained the ratio of the total area under the curve which is produced by the thermal effects of the dissociation of the $MgCO_3$ to the total area under the curve which is produced by the thermal effects of the dissociation of the $CaCO_3$ in the structure. Then, assuming 0.60 as the value of this ratio for dolomite, the value of the ratio was calculated for huntite and for a mixture of 75 per cent magnesite and 25 per cent dolomite, specimen *W*-199. The concordance between the measured and calculated value of the ratio is excellent. It must be emphasized that the above calculations are based on the arbitrary assumption that the area under the curve for the thermal effects produced in the dissociation of magnesite are not greatly different from those produced in the dissociation of the $MgCO_3$ in the structures of huntite and dolomite.

Kelley and Anderson (1935) give equations for calculating the heat of the dissociation of calcite and of magnesite. The values of ΔH for the two-stage dissociation of the dolomite from Millville, West Virginia (curve *C*), were calculated by means of these equations and the measured temperature of the two maxima, 792° C. and 926° C. The calculated values are $\Delta H_{1065} = 23{,}440$ calories for the dissociation of the $MgCO_3$ in the structure and $\Delta H_{1199} = 39{,}702$ calories for the $CaCO_3$. The ratio of these values of ΔH is 0.590, and this is in excellent agreement with the value 0.584 obtained by areal measurement.

CHEMICAL PROPERTIES

(1) *Pyrognostics*

Huntite is infusible before the blowpipe. A fragment, when held in the forceps and heated, glows and colors the flame a rich red. The heated fragment, when dropped onto wet turmeric paper, turns the paper brown, thus showing an alkaline reaction. Fragments dissolve quickly and with brisk effervescence in cold (1:1) acids such as HCl, HNO_3, or H_2SO_4. Gypsum is precipitated when H_2SO_4 is used as the solvent. Huntite does not become colored with Meigen's solution.

(2) *Chemical analysis and composition*

A chemical analysis was made on pure material, specimen *A*-1007a, from the Ala-Mar deposit, and the results are given in Table 4.

TABLE 4. ANALYSIS OF HUNTITE FROM THE ALA-MAR DEPOSIT, NYE COUNTY, CURRANT CREEK, NEVADA

	Per Cent	Mol. Numbers	Mol. Ratios
MgO	34.09	0.8455	3.07
CaO	15.42	0.2750	1.00
MnO	.001		
H_2O-	.47		
H_2O+	.86		
CO_2	48.85	1.1102	4.03
SiO_2	.06		
Al_2O_3	none		
Fe_2O_3	none		
FeO	none		
TiO_2	none		
SO_3	none		
BaO	none		
Sum	99.75		

Analyst: R. E. Stevens

This analysis yields the formula $3MgO \cdot CaO \cdot 4CO_2$ or $3MgCO_3 \cdot CaCO_3$ or $Mg_3Ca(CO_3)_4$. This composition lies between dolomite and magnesite and is in the ratio of two moles of magnesite to one mole of dolomite.

K. J. Murata made a spectrographic examination of huntite obtained from a cavity filling in specimen no. *N.M.186* from the Nevada Magnesite deposit; this examination showed that the minor constituents present in amounts of a few hundredths of a per cent, or less, were Si, Fe, Sr, Ba, Ti, B, Al, and Mn.

NOMENCLATURE

Huntite is named in honor of Professor Walter Frederick Hunt, sometime chemist in the Chemical Laboratory, U. S. Geological Survey, for many years professor and head of the Mineralogical Laboratory at the University of Michigan, and, since 1922, editor of this Journal. The author is particularly happy to name the mineral after one of his former teachers in the Graduate School of the University of Michigan, as a token of appreciation to a professor who was everything that he should be to a graduate student.

ACKNOWLEDGMENTS

During the latter part of 1942 the author was requested by Eugene Callaghan to undertake the study of the mineralogy of the magnesite deposits of the Currant Creek area. The original specimens of huntite were discovered in collections made by C. J. Vitaliano and E. Callaghan. These were supplemented in 1944 by specimens collected by the author and Callaghan. The author is indebted to R. E. Stevens for the chemical analysis, to J. J. Fahey for the determination of the density, to E. J. Dwornik for the electron micrographs, and to K. J. Murata for spectrographic data. J. M. Axelrod made preliminary x-ray examinations of this material and confirmed its unique character. To all of these colleagues the author expresses his sincere thanks.

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STUDIES IN THE MICA GROUP; MINERALOGY OF THE ROSE MUSCOVITES*

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Abstract

Rose muscovite is a variety of muscovite with distinctive paragenetic, optical, and chemical characteristics. It occurs in replacement units of complex pegmatites with a hydrothermal Na-Li phase and is the youngest mica to form, even following lepidolite. Indices of refraction are lower than those of normal muscovite, with average values: $\alpha=1.555$, $\beta=1.586$, $\gamma=1.592$. Chemically the rose muscovites are characterized by the consistent presence of small amounts of Fe_2O_3, MnO, Li_2O, Rb_2O and F. The unusual color is not related to the Li_2O content but depends upon the essential absence of Fe^2 and the dominance of Mn over Fe^3.

Introduction

This paper purposes to describe the mineralogy and paragenesis of the unusual rose or pink pegmatitic muscovites and to discuss the interrelation of their chemical composition and color. The field work was done by the senior author in New Mexico and Colorado at various times during the summers of 1948, 1949, 1951, and 1952, and in Finland, Sweden, and Norway during the summer of 1950. Laboratory investigations were conducted jointly in 1951–1952.

The European field studies were assisted by a grant from the Faculty Research Fund of the Horace H. Rackham School of Graduate Studies, University of Michigan, which fund also paid the cost of five new chemical analyses. This study is a part of the research program, Natural Mica Studies, sponsored by the U. S. Army Signal Corps, Squier Signal Laboratory, Ft. Monmouth, New Jersey, under the general supervision of Dr. S. Benedict Levin and administered as Project M-978 of the Engineering Research Institute, University of Michigan. The writers are indebted to Charles H. Hewitt and Donald W. Levandowski of the Department of Mineralogy for much valuable laboratory assistance and to the following for gifts and loans of specimens: Professor Clifford Frondel, Harvard University; Professor Horace Winchell, Yale University; the late Dr. H. V. Ellsworth, Canadian Geological Survey; Dr. Harald Björlykke, Norwegian Geological Survey; Dr. Frans E. Wickman, Royal Museum of Stockholm; Mr. R. Jacobson, Nigerian Geological Survey;

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* Contribution from the Department of Mineralogy and Petrography, University of Michigan, No. 169.

and Dr. H. P. Rowledge, Government Chemical Laboratories, Western Australia.

OCCURRENCES

United States

New England

Goshen, Massachusetts: The occurrence of rose mica at Goshen was recorded by Hitchcock (1841, p. 701). Apparently the first reference to the possibility that this rose mica was not lepidolite was given by Dana (1850), who states (p. 361), ". . . a rose mica is obtained at the albite vein in Chesterfield, Mass., and at Goshen in the same State. . . . The Goshen mineral is of difficult fusibility and slight lithia reaction, and may not be of this species." Mallet (1857) analyzed the Goshen mica and confirmed its very low lithia content, concluding it was muscovite.

The Goshen pegmatites have been described by Julien (1879*A*, 1879*B*), Emerson (1898, 1917), Browne (1933), and Hess and Ralston (1938). The mineral assemblage includes albite, spodumene, blue, green, and pink tourmaline, colorless and light yellow beryl (goshenite), columbite, and cassiterite. The rose mica is associated with the pink tourmaline. Because of its reported large 2E, it is possible that the rose mica from nearby Chesterfield also may be muscovite, but specimens were not available for examination.

Bolton, Massachusetts: The Bolton marble quarry, famous for lilac scapolite, has been described by Palache and Pinger (1923) who state (p. 154), "There is an abundance of coarse granite pegmatite boulders in the immediate neighborhood of the quarry but the rock could not be discovered in place . . . the formation of scapolite on the unusually large scale . . . is due to the action of pegmatite intrusions upon the materials of the gneiss limestone contact zone."

Specimens of rose mica-bearing pegmatite from Bolton show the following megascopic zonal arrangement:

(*a*) Wall rock, a ¼-inch remnant of finely granular quartzose schist.
(*b*) Border zone, ½-inch thick, fine-grained white feldspar-quartz rock with minor specks of schorl. The gray quartz rods tend to be normal to the wall.
(*c*) Coarse-grained cleavelandite-smoky quartz-muscovite pegmatite, with mica books as large as two inches across and a few schorl crystals.

A thin section of the border zone shows a fine-grained intergrowth of orthoclase with poikilitic quartz, oligoclase with a narrow sodic selvage, quartz, and rounded deep blue tourmaline grains.

In a thin section of unit *c*, cleavelandite blades can be seen to replace orthoclase of an anhedral equigranular quartz-orthoclase intergrowth. Both the earlier minerals are minutely fractured and locally show marked wavy extinction, neither of which characterizes the albite. Some of the cleavelandite is veined by later quartz and is associated with ragged crystals of irregularly mottled blue tourmaline. Nearly all the mica books have combined wedge and fishtail structure, but some books include flattened euhedral schorl crystals. Some of the smaller books are entirely rose colored, but most are mainly pale yellowish green with rims of pink along one side (Fig. 1). The colors intergrade. Several larger wedged books vary in the proportions of the two colors in different sheets, with green predominating at one end of the book and rose at the other.

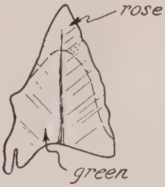

FIG. 1. Zoned rose and green muscovite from Bolton, Massachusetts. ×½.

Haddam Neck, Connecticut: The pegmatite at Haddam Neck contains zoned mica crystals, quartz, albite, and microcline together with green and red tourmaline, green and red apatite, green and brown fluorite, colorless to pink beryl, cookeite, lepidolite, columbite, and microlite (Bowman, 1902, 1903). The columnar mica crystals, as much as 3 inches across and about 2 inches long, are composed of a core of green-white muscovite and a rim of lepidolite, which is in turn coated by fibrous rose muscovite. This fibrous overgrowth consists of parallel *c*-axis needles of rose muscovite. The cleavage plates are made up of minute rhombic units intergrown in a 60°–120° mosaic. The lepidolite-rose muscovite contact is sharp and straight, and the rhombs are either in parallel position or in twin position (Fig. 2). Bowman (1902, p. 120) states, "Among (the micas) themselves they never show any deviation from the order, greenish-white muscovite—lepidolite—fibrous muscovite." Bowman notes the occurrence of similar material at Auburn, Maine, which could not be verified by the writers.

Lord Hill pegmatite, Rumford, Maine: According to Woodard (1951) the Lord Hill pegmatite contains an aplitic border zone of oligoclase, myrmekite, microcline, biotite, and muscovite, a wall-zone of albite, quartz, microcline-perthite, and muscovite and a quartz-perthitic microcline core. Aggregates of cleavelandite, fluorite, spessartite, topaz, and muscovite form replacement bodies in the core. Light pink muscovite is interstitial to the cleavelandite blades.

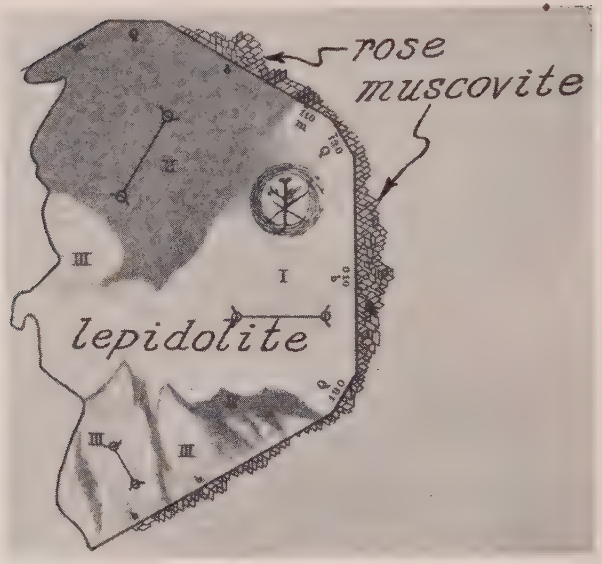

FIG. 2. Oriented overgrowth of fine-grained rose muscovite on lepidolite, after Bowman (1902A). ×5.

Virginia

Rutherford pegmatite, Amelia Court House: Glass (1935) describes a shell-pink variety of muscovite that occurs as tiny fan-shaped plates in the angular cavities between interlocking platy cleavelandite blades. Of five varieties of sericite studied by her, two (varieties 3 and 4) vary in color from aniline lilac to grayish lavender. These (p. 755) ". . . occur in two forms—waxy, talc-like masses, which under high magnification, appear to be aggregates of minute scales or fibers. . . . Varieties 3 and 4 are optically identical. These varieties of muscovite are found intimately associated with the bluish colored cleavelandite albite, in some places filling interstices between the albite plates." Glass also notes that a green sericite (Variety 1) ". . . is penetrated by veinlets of the lilac-colored variety with cleancut contacts."

New Mexico

Harding pegmatite, Taos County: The Harding pegmatite has been known as a source of fine and unusual pegmatitic mineral specimens for several decades, and commercial quantities of lepidolite, spodumene, beryl, tantalite, and microlite have been produced from it. Schaller and Henderson (1926), Hirschi (1928), Hess (1933), Just (1937), Northrop (1942), Jahns and Wright (1944), Montgomery (1950) and Jahns (1951) have described its mineralogy and geology, and Roos (1926), Soulé

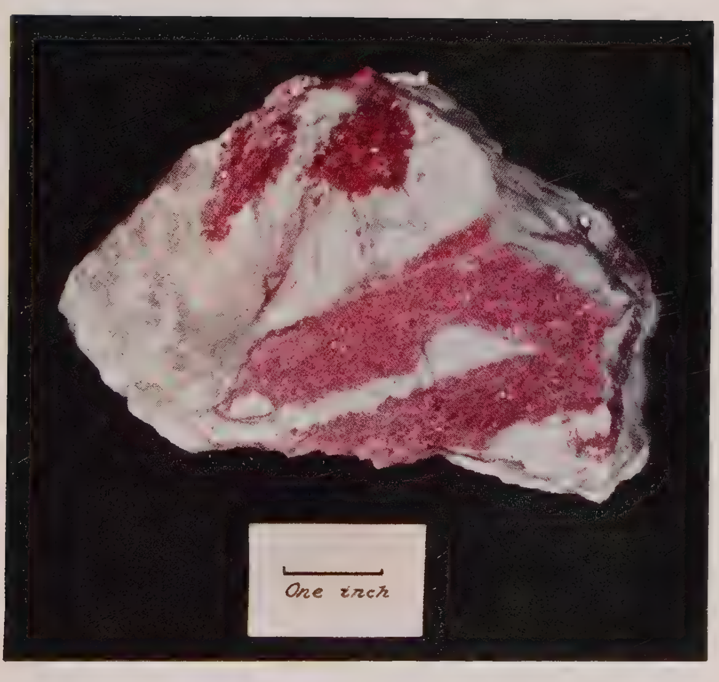

PLATE I. Rose muscovite pseudomorphous after spodumene, Harding pegmatite, New Mexico.

(1946) and Berliner (1949) have emphasized the economic aspects of the deposit.

The Harding pegmatite is a nearly horizontal lens-like dike, 50–60 feet in average thickness, which has been traced southwestward down-dip for 1,000 feet. It is known to contain concentrations of beryl, lithium and tantalum minerals for distances of 350 feet along the strike and 650 feet down-dip. The dike is markedly layered:

(0) Hanging-wall amphibolite.
(1) Quartz-muscovite-albitized microcline with minor apatite, columbite, lepidolite; white to rose beryl very abundant locally; 6 inches to 5 feet thick. A similar zone lies locally above the footwall.
(2) Massive quartz, 6 feet thick.
(3) Quartz with long slender spodumene laths; some microcline and beryl near the base; 20 feet thick.
(4) Coarse-grained spodumene-microcline-quartz ("spotted rock"); variable amounts of albite, muscovite, lepidolite, tantalite, and brown microlite; 30 feet thick.
(5) A series of discontinuous lepidolite pods containing spodumene, smoky quartz and yellow microlite.
(6) Cleavelandite-rose muscovite; 20–30 feet thick.
(7) Quartz-cleavelandite; as much as 15 feet thick.
(8) Sugary albite rock; 10 feet thick.

This complete sequence is not present in all parts of the dike, for some units are lensoid and discontinuous. Most of the rose muscovite is concentrated in the cleavelandite-rich replacement unit (6) below the "spotted rock" central unit (4), but some pods of cleavelandite-rose muscovite rock also have been localized along the contact between the quartz-lath spodumene zone (3) and the overlying massive quartz zone (2).

The cleavelandite-rose muscovite rock shows spectacular color contrasts (Plate I). Much of the rose mica occurs as aggregates of fine-grained flakes ($\frac{1}{4}$-inch or less in diameter) that form thin tablets 1–2 inches wide and 5–6 inches long, pseudomorphous after spodumene crystals. All stages of replacement are present, from spodumene laths coated by rose mica films or encroached by a network of ramifying mica veinlets, through composite laths of ragged spodumene relicts and rose mica in varying proportions, to rose mica-granular quartz laths containing only a few rounded blebs of spodumene or none at all (Fig. 3). A few small flakes of yellow microlite and greenish-blue apatite also occur in some of the pseudomorphs. The muscovite flakes are generally rudely oriented with (001) parallel with the flat direction of the lath (100) of the spodumene. Rose muscovite also occurs as elongated to rounded pods, several inches across, of randomly oriented flakes filling interstices between the cleavelandite blades and rosettes (Fig. 4). A few similar mica pods occur in smoky quartz. Much rarer are fist-sized, curved masses of rose mica in which the grain size ranges from cryptocrystalline to coarse-grained.

Locally rose muscovite is associated with anhedral to platy white beryl. Through the kindness of Dr. W. T. Schaller the senior writer was able to examine an apparently unique specimen, collected by Professor Richard H. Jahns, which consists of very fine-grained scaly, rose mica pseudomorphous after a single stubby crystal, 1¼ inches on edge and exposed for three inches along the *c*-axis direction enclosed in massive pink beryl. The crystal is orthorhombic with prisms, basal pinacoid and modifying dome faces and could have been either topaz or andalusite from the habit; from the association topaz seems more probable.

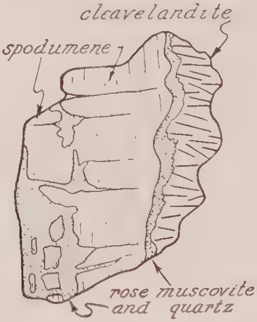

FIG. 3. Rose muscovite replacing spodumene, Harding pegmatite, New Mexico, ×½.

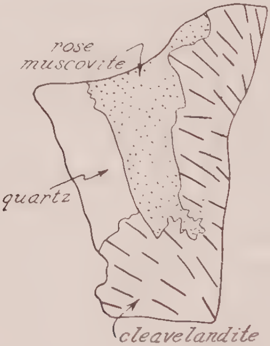

FIG. 4. Rose muscovite pod along quartz-cleavelandite contact, with minor replacement of cleavelandite, Harding pegmatite, New Mexico. ×½.

Studies of thin sections of the cleavelandite-rose mica rock support the megascopic paragenetic interpretations. Spodumene blades are invaded by rose muscovite and quartz. Veinlets of mica flakes also follow along contacts between cleavelandite blades or between cleavelandite and quartz, corroding the albite, and bisect masses of anhedral beryl. Many of the muscovite flakes are zoned with a very thin rim that displays a lower birefringence than that of the main central part of the crystal.

Jahns (written communication) states, ". . . the several specimens that I do have and the several other observations that I made in the field all indicate that the pink muscovite is later than the gray and purple lepidolite. In all these places the lepidolite is veined by the muscovite—the pink muscovite, that is. There also is a little fine-grained gray to greenish gray muscovite in the Harding pegmatite that is later even than the pink muscovite."

These combined observations indicate that the rose muscovite is one of the very latest minerals to form. It followed spodumene, cleavelandite, alkali beryl and lepidolite. The lepidolite itself is younger than microcline and the coarse silvery-green book muscovite.

Pilar pegmatites, Taos County: The Pilar purple muscovite, described by Schaller and Henderson (1926), differs markedly from the Harding rose muscovite, and all other muscovites, in composition, optical properties, and paragenesis. It occurs in simple, small quartz-microcline pegmatite dikes, and none of the minerals typical of other rose muscovite occurrences are found with it.

Pittlite pegmatites, Mora County: Several lithium-tantalum pegmatites on the eastern side of the Sangre de Christo Range near Rociada were examined during June 1951. Brief descriptions of these by Jahns (1946*B*) and Page (1950) do not specifically mention the rose muscovite that is relatively abundant in one of the dikes. Two elongated pegmatite pods, cutting the foliation of fine-grained amphibole schist, strike slightly east of north and are exposed over a distance of about 100 feet. The northern body is 50 feet long, 16 feet wide and is exposed to a depth of 40 feet. Its western contact dips steeply west-northwest, whereas the eastern wall dips about 60° east-southeast. The other dike, a few feet to the south, is 10 feet wide and is exposed for 45 feet.

The northern pegmatite is zoned with:

(1) Wall zone of quartz-sodic plagioclase-muscovite.
(2) Core of quartz with some microcline.
(3) A unit along the western margin of the core consisting mainly of quartz, cleavelandite and lepidolite, with a maximum thickness of about 10 feet, tapering southward.

The southern dike contains similar units, except that the quartz-cleavelandite-lepidolite rock occurs as two irregular central pods. Rarer minerals occurring in the cleavelandite unit are tourmaline, beryl, topaz, spodumene, amblygonite, apatite, monazite, microlite, betafite, tantalite, fluorite, and bismutite.

As in the Harding dike, the rose mica is associated closely with cleavelandite, which at the Pittlite deposit is mottled white to blue-gray. Much of it is interstitial to the blades as elongated or irregular pods, usually one inch or less across. The microscope shows veinlets of muscovite across cleavelandite blades or along blade contacts. Many mica flakes have a thin rim with slightly lower birefringence. Anhedral white beryl also is cut and dactylicly penetrated by mica veins. Many mica flakes are bent or crenulated, with undulatory extinction. Some are peripherally replaced by an aggregate of exceedingly fine-grained sericite of slightly lower refractive index than the rose muscovite. Associated are grains of deep brown microlite and coarser flakes of purple lepidolite. Fist-sized masses of milky topaz are enclosed in a jacket of very fine-grained waxy to coarser flaky rose muscovite. The rose muscovite is younger than cleavelandite, beryl, topaz, and probably lepidolite, though for the latter the relations are not clear-cut.

Petaca District, Rio Arriba County: Rose muscovite has been found in pegmatites of the Petaca district, whose geology and mineralogy have been studied by Just (1937), Northrop (1942), Heinrich and Jahns (1946), Jahns (1946*A*) and Wright (1948). A complete bibliography of the literature on these deposits has been compiled by Jahns (1946*A*, pp. 15–16). Most of the pegmatites of economic importance are well zoned and also commonly contain superimposed hydrothermal replacement units of cleavelandite and muscovite. The chief accessory minerals of these replacement bodies are fluorite, columbite, monazite, samarskite, bismutite, and copper sulfides. Pink muscovite was observed in eight deposits, or 12% of the 69 deposits studied by Jahns. The main occurrences are in the Apache and Globe pegmatites, others are in the Kiawa, Canary Bird, Cribbenville, Conquistador and Fridlund deposits. In none, however, is the rose mica abundant. It appears typically as small irregular pods, six inches or less long, in cleavelandite or sugary albite or in quartz pods within cleavelandite units. Flakes are rarely larger than ¼-inch. Columbite and small flakes of pale apple-green muscovite are associates, and in one pod from the Apache mine, pink muscovite grades into green muscovite. Wright's conclusion (1948, p. 688) appears to apply generally to the district, ". . . the pink muscovite is also one of the latest minerals in the Globe pegmatite."

Colorado

Quartz Creek District, Gunnison County: The complex pegmatites near Ohio City resemble the Harding and Pittlite dikes, chemically and mineralogically. Eckel (1933), Landes (1935), Fairchild et al. (1935), Hanley (1947), Hanley, Heinrich and Page (1950) and Staatz and Trites (1950) have presented descriptions of the geology and mineralogy of these pegmatites. At the Brown Derby mine are exposed three parallel dikes that strike N. 56° E. and dip 30–35° SE. The Brown Derby No. 1 dike, the largest and mineralogically most complex, is just over 900 feet long, as much as 40 feet wide at the surface and has been explored down dip for 65 feet.

The lithium-rich part of the dike is strikingly layered:

(0) Footwall, hornblende schist.
(1) Fine-grained albite-quartz unit, with local cleavelandite and accessory schorl, beryl, and lepidolite; average thickness 1.5 feet.
(2) Cleavelandite-quartz-lepidolite-topaz unit with accessory beryl and alkali tourmaline; books of deep lavender lepidolite as much as 10 inches across form a distinct band at the top of the unit; average thickness 2 feet.
(3) Lepidolite-quartz-cleavelandite-microlite unit. The lepidolite is generally fine-grained; alkali tourmaline is an accessory mineral; maximum thickness 8 feet.
(4) Cleavelandite-quartz-curved lepidolite unit with local concentrations of alkali tourmaline; maximum thickness a little less than 2 feet.
(5) Quartz-cleavelandite unit. Mainly massive quartz with veins and replacement masses of cleavelandite; 4 feet thick.
(6) Hanging-wall, hornblende schist.

North of the lithium-rich part the dike consists mainly of quartz-microcline-albite-muscovite rock with minor schorl and beryl. South of the lithium-rich part the dike forks into an eastern limb consisting of quartz-microcline-albite-muscovite pegmatite and a western branch made up mainly of cleavelandite-quartz rock with some lepidolite and alkali tourmaline. In one pit in the western limb there is exposed on the hanging-wall contact a pod of fine-grained albite-quartz-zinnwaldite pegmatite with considerable columbite, monazite, schorl, garnet, and minor gahnite and fluorite.

Most of the rose muscovite appears in unit 2 and is closely associated with the topaz, which occurs in milky, rounded or rudely-faced crystals, as much as four feet long and one foot in diameter. The rose mica, which forms a jacket around the topaz crystals, coating and corroding them, varies from microscopically felted aggregates to coarse flakes $\frac{1}{2}$ inch across. From many of the coatings veinlets of coarse flakes extend into and through the topaz crystals (Fig. 5). Some of the topaz crystals are light pink in color but show no megascopic mica. Under the microscope, these are seen to be strongly brecciated in diverse directions on a small scale. Veinlets of very fine-grained pink mica follow the brecciated zones, surrounding and replacing randomly oriented topaz fragments.

The White Spar No. 1 and No. 2 pegmatites (Hanley, Heinrich and Page, 1950, pp. 77–80) are about 0.8 mile northeast of the Brown Derby group. The White Spar No. 1, which is about 200 feet long and as much as 85 feet wide, trends N. 20° E. and dips 30–35° SE. A wall zone of medium-grained quartz-microcline-muscovite pegmatite surrounds a quartz core four feet thick. A core margin replacement unit of cleavelandite-lepidolite rock, six feet thick, has been developed along the wall zone-core contact, together with a small pod of lepidolite-microlite pegmatite on the hanging wall side of the core. In the core margin unit also occur beryl, topaz, alkali tourmaline, microlite, columbite and rose muscovite. The rose mica occurs in several ways:

(1) As fine to coarse-grained veinlets in and peripheral replacements of topaz.
(2) As veinlets in cleavelandite, with flakes as much as one inch across.
(3) As fine-grained aggregates interstitial to wedge-shaped cleavelandite clusters.
(4) As minute aggregates of flakes in quartz-microcline-oligoclase pegmatite along the outer edge of the core-margin unit. Here coarser flakes occur in quartz, whereas very fine-grained vermicular patches replace microcline.

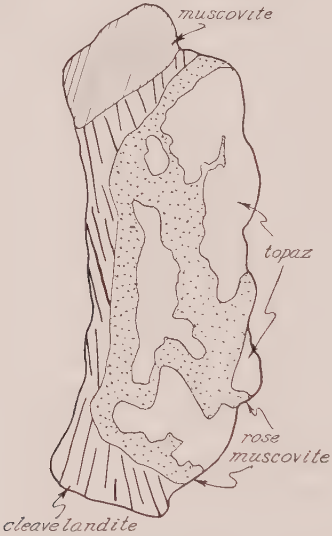

Fig. 5. Rose muscovite coating and veining topaz crystal. Brown Derby No. 1 pegmatite, Colorado. ×½.

California

Mesa Grande: An analysis is recorded of a pink muscovite from Mesa Grande (Clarke, 1915, p. 330). Other minerals present in this deposit include alkali tourmalines, alkali beryl, bavenite, apatite, pollucite, cleavelandite, and lepidolite.

Canada

Manitoba

Pointe du Bois: Rose muscovites occur in several deposits in the Pointe du Bois area, particularly in the Bear and Annie pegmatites (Ellsworth, 1932; Stockwell, 1933*A*, 1933*B*). The Bear pegmatite contains three well-defined units:

(1) Lower: Medium-grained albite-quartz-microcline-pegmatite, overlain by aplitic scallopy-banded albite-quartz-garnet rock ("line-rock") and above that a 3–6-inch cleavelandite band. Small amounts of lepidolite, zinnwaldite, and spodumene occur in the "line-rock."
(2) Middle: Lower and upper bands, containing large microcline crystals and quartz-spodumene crystals with interstitial quartz, cleavelandite, and lepidolite occur around an inner layer of lepidolite-cleavelandite-quartz rock with local beryl, spodumene, and rose or purple muscovite.
(3) Upper: Quartz-muscovite pegmatite with minor microcline separated from unit 2 by a one-foot cleavelandite band.

Light lilac muscovite forms a massive rock of very fine scales, 1 mm. or less in diameter, in the inner layer of unit 2. It was found exposed in an area 5–10 feet across, together with quartz, cleavelandite and smaller amounts of spodumene, topaz, white to pink beryl, montebrasite, and lithiophilite. Lilac curved muscovite, with 1.80% Li_2O, is associated with gray curved muscovite locally in the same unit.

Quebec

Fiedmont area, Abitibi County: Tremblay (1950) has described pegmatites in the Fiedmont area that have quartz cores enclosed in spodumene-rich units also containing quartz, microcline, cleavelandite, lepidolite, and accessory sugary albite, beryl, spessartite, columbite-tantalite, microlite, betafite, bismuthinite, molybdenite, and powellite. Both green and buff spodumene may be partly or completely altered to a lithium-bearing mica. The relpacement begins from the margin or from fractures. According to Tremblay (private communication) this mica is pink in color, and the lithium content was assumed from the color and the association. It has the optical properties of muscovite and forms rosettes or platy aggregates.

Europe

Norway

Iveland-Evje District, Setesdal: The pegmatites of the Iveland-Evje district have been investigated by Björlykke (1935*A*, 1935*B*, 1936, 1937, 1939) and by Barth (1928, 1931). Deposits from which rose muscovite has been recorded are Skripeland No. 1, Birkeland No. 2 and 3 and Landsverk. These are pegmatites in which occur a strongly developed cleavelandite phase and various combinations of accessory spessartite, topaz, beryl, schorl, microlite, tantalite, zircon, gadolinite, monazite, xenotime, thorite, uranothorite, betafite, allanite, fergusonite, euxenite, and bismuthinite. The rose muscovite is closely associated with a green muscovite and with cleavelandite, occurring as:

(1) A thin layer between topaz and cleavelandite, with the cleavage normal to the topaz surface.
(2) Interstitial to cleavelandite blades.
(3) In vugs in cleavelandite with small albite crystals.

Flakes as large as one-half inch in diameter occur, but most are smaller.

Höydalen, Tördal, Telemark: A cleavelandite-rich pegmatite at Höydalen contains an albite-quartz-spessartite outer zone around an amazonite core largely replaced by cleavelandite and quartz with accessory gadolinite, alvite, yttrotantalite, cassiterite, topaz, fluorite, lepidolite, lithium muscovite, and both bladed and massive pink muscovite (Oftedal, 1944). Assuming that the Sc content of micas decreases with the temperature (and thus time) of their formation, Oftedal (1943) has determined the rose muscovite to be the youngest of the three micas:

	g./ton Sc	% Li_2O
(1) Gray lithium-rich muscovite	700	1.5
(2) Red violet lepidolite	5	5.0
(3) Pink muscovite	<3	<0.1–0.3

The rose muscovite accompanies cleavelandite, with relicts of gray albitized microcline appearing in some specimens. In thin section the gray lithium-rich muscovite, which also is mainly interstitial to cleavelandite, replaces the feldspar marginally and not uncommonly is separated from the plagioclase by a narrow reaction rim of fine-grained quartz and sillimanite needles. Small anhedral specks of fluorite also occur in the albite.

Sweden

Varuträsk: The minerals of the complex pegmatite at Varuträsk have been described in numerous papers by Quensel and his students. Quensel (1952), in his description of the paragenesis, lists a complete bibliography. Berggren (1940, 1941) has analyzed the micas, and Lundblad (1942) has studied them optically. Berggren (1940) states that the very fine-grained rose mica occurs in the eastern wing of the pegmatite, which is a trough-shaped body whose axis trends north-northwest. The zones of the pegmatite include (Quensel, 1952):

(1) Fine-grained quartz-muscovite border zone.
(2) Wall zone of coarse-grained quartz, muscovite, and schorl.
(3) Outer intermediate zone of albite, microcline, quartz, and beryl.
(4) Inner intermediate zone of microcline, quartz, muscovite, spodumene, and amblygonite.
(5) Massive quartz core.

Upon these zones several hydrothermal replacement units have been developed:

(6) A high-temperature lithium unit with lepidolite, spodumene, quartz, petalite, and alkali tourmaline.
(7) A high-temperature cesium unit of pollucite and quartz.
(8) An intermediate- to low-temperature unit of cleavelandite, quartz, lepidolite, blue tourmaline, Mn-Fe phosphates, Cb-Ta minerals, uraninite, and cassiterite.
(9) Veins of fluorite.

The detailed paragenesis of the rose muscovite is not fully known, but it occurs in the cleavelandite replacement unit (8). Another rose muscovite, described as a purple oncosine with 1.10% Li_2O is found in veinlets in the eastern limb of the deposit.

Finland

Kimito District: According to Pehrman (1945), the large muscovite books of the Kimito pegmatites are yellow or gray in color, whereas the muscovite that occurs in fine scaly masses is violet or green. He lists the optical constants of violet muscovite from the Lemnäs pegmatite.

Africa

Nigeria

Ejiba: Jacobson and Webb (1946, p. 20) state, "An unusual type of muscovite was collected from an intensely albitized microcline-quartz-muscovite pegmatite near Ejiba. The muscovite occurs in crystals elongated along the *c*-axis. The books are about half an inch in diameter and up to three inches long. At one end they are apple-green in colour and at the other lilac. The colours grade into one another along the length of the crystal."

The color arrangement, variation and intergradation in this mica are very similar to those in material from Bolton, Massachusetts. The albitized pegmatites contain the accessory suite: cleavelandite, sugary albite, lepidolite, alkali tourmaline, apatite, monazite, triphylite, amblygonite, beryl, cassiterite, columbite-tantalite, tapiolite, and microlite.

Australia and Asia

Western Australia

Pilbara, North-West Division: The Strelley pegmatite (Ellis, 1950) contains scattered masses of pale violet to purplish muscovite, cryptocrystalline to massive and translucent. The mica replaces (potash?) feldspar along with pale purple chalcedony. In the Pilgangorra deposits occur numerous small lenses and bunches of scaly pink muscovite, associated with albitized parts of the pegmatite. Spodumene in these pegmatites shows varying degrees of alteration to purplish pseudomorphs composed mainly of fine-grained muscovite. Partial analyses of these and numerous other rose muscovites from Wodgina and West Wodgina have been made by Rowledge (1945). Simpson (1927) also reports a rose muscovite from Londonderry. It varies from colorless to pale mauve, lilac or sea green and occurs in association with lepidolite. Spodumene in pegmatites in the McPhees Range (Simpson, 1938) has been replaced similarly by gray to nearly purple muscovite.

Japan

Suizawa, Ishigura, Mi-e Prefecture: A rose muscovite from Suizawa has been analyzed by Iimori and Yoshimura (1929).

GENERAL PARAGENESIS

Although the data are incomplete for some of the deposits, the previous descriptions are sufficient to establish several generalizations regarding the paragenesis of the rose muscovites:

(1) Rose muscovites apparently are restricted to replacement units in zoned pegmatites.
(2) The replacement units are of the Na-Li type, commonly containing cleavelandite, lepidolite, spodumene, topaz, beryl, and tantalum minerals.
(3) Rose muscovite forms partial or complete pseudomorphs after spodumene and topaz, also replaces cleavelandite and beryl and occurs as overgrowths on lepidolite or normal muscovite. Thus, in general, rose muscovites are among the very youngest of the minerals of the replacement unit, and the sequence of mica formation is: muscovite, lepidolite, rose muscovite.

This conclusion opposes that of Stevens and Schaller (1942, p. 533) who state, "In the full development of the lithium stage, the mica formed is essentially lepidolite. No new muscovite is formed at this stage. The muscovite now present in specimens from the lithium phase is residual muscovite from the preceding sodium phase, as evidenced by the many examples of muscovite completely or partially changed to lepidolite, by the pink color of this muscovite with percentages of Li_2O intermediate between those of the greenish muscovites of the albite phase and those of lepidolite, and by the border of lepidolite formed around the muscovite, which is often colored pink adjacent to the lepidolite border."

It appears that once the peak of lithium replacement has passed, muscovite of the rose type is formed, either by direct precipitation along fractures and in cavities or through replacement of a lithium mineral such as spodumene and of non-lithium minerals such as topaz and cleavelandite.

PHYSICAL PROPERTIES

Color

Exact color determinations of rose muscovites are considerably hampered by the physical nature of most of the specimens. Because of the general fine grain and the very pale colors, color determinations on single sheets of standard thickness are not possible. Color can only be determined for aggregates, which under standard illumination show some variations in their tints owing to:

(1) Variations in grain size.
(2) Variations in thickness of specimens.
(3) Variations in amount and type of admixed material or in underlying matrix material.

However, despite these variables, reproducible determinations were obtained by rectangularly masking the specimens and correlating the exposed areas in juxtaposition with units of the Ridgway (1912) tables.

Except for determinations by Schaller and Henderson (1926), color designations of rose muscovite have been entirely qualitative, employing such terms as pink, rose, magenta, lilac, lavender, purplish, purple, and violet. Our color designations are listed in Table 1. In thin section the rose muscovites are colorless. Schaller and Henderson (1926, p. 10) note very weak pleochroism in thick scales of the Harding mica. There is normally no difficulty in distinguishing, colorimetrically, between the rose muscovites and the so-called "ruby" muscovites. As has been pointed out by Jahns and Lancaster (1950, p. 43), most "ruby" micas are buff, drab, and cinnamon brown. However, because of their color, the rose muscovites have been confused repeatedly with lepidolites. Wherever the two similarly colored micas are closely associated, the lepidolite generally is characterized by a more bluish or purplish tint (Schaller and Henderson 1926, p. 9), but in some cases color alone does not suffice and an Li_2O determination or an x-ray photograph is required. Because of this color similarity probably many rose muscovites have remained unrecognized, and the variety may well be more common than the published record indicates.

Optical Constants

In Table 1 also are listed the optical constants of representative rose muscovites studied as well as some scattered data from the literature. The ranges in values are remarkably narrow in our measurements:

	Average values
$\alpha=1.553$–1.556	$\alpha=1.555$
$\beta=1.584$–1.595	$\beta=1.586$
$\gamma=1.587$–1.599	$\gamma=1.592$
$2V=42$–$46\frac{1}{2}°$, $r>v$ weak	$2V=45°$

All three indices were measured on an Abbé refractometer, and β and γ also were checked by means of the immersion method. 2E was determined by means of the Mallard equation, and 2V was calculated.

TABLE 1. OPTICAL PROPERTIES OF REPRESENTATIVE ROSE MUSCOVITES

Locality	Ridgway Plate No. and Color	α	β	γ	Axial Angle	Reference
Goshen, Mass.	—	—	—	—	2E=74–76°	Mallet, 1857
Bolton, Mass.	XXVI, cameo pink	1.556	1.590	1.595	2V=45°	This paper
Haddam Neck, Conn.	—	—	—	—	2E=75½°	Bowman, 1903
Amelia, Va.	—	1.553	1.582	1.588	calc. 2V=48°	Glass, 1935
Amelia, Va.	—	1.552	1.583	1.590	2V=45°	Glass, 1935
Harding, N. M.	XXVI, rosalane purple rosalane pink XXXVII, eupatorium purple daphne pink	1.558	1.587	1.591	2E=69° calc. 2V=42°	Schaller and Henderson, 1926
Harding, N. M.	XII, deep rose pink	1.555	1.587	1.592	2V=45°	This paper
Harding, N. M.	XXXVIII, pale Persian lilac	1.555	1.587	1.592	2V=46½°	This paper
Pittlite, N. M.	XXVI, cameo pink	—	1.586	1.589	2V=46°	This paper
Apache, N. M.	XII, rose pink	1.555	1.586	1.592	2V=43½°	This paper
Globe, N. M.	XXVI, cameo pink	—	1.588	1.592	2V=44°	This paper
Brown Derby, Colo.	XXVII, pale vinaceous	1.553	1.584	1.587	2V=45°	This paper
Brown Derby, Colo.	XXVII, pale vinaceous	1.555	1.588	1.591	2V=45°	This paper
White Spar No. 1, Colo.	XXVII, pinkish vinaceous	1.553	1.586	1.590	2V=45–46½°	This paper
Iveland, Norway	XXVI, cameo pink	1.554	1.586	1.590	2V=45°	This paper
Höydalen, Norway	XXXVIII, pale rhodonite pink	1.553	1.585	1.590	2V=45°	This paper
Höydalen, Norway	XXVII, livid pink	1.555	1.586	1.592	2V=46½°	This paper
Lemnäs, Finland	—	—	1.589	1.594	2V=44°56′	Pehrman, 1945
Ejiba, Nigeria	XXXIX, pale brownish vinaceous	—	1.587	1.592	2V=42°	This paper
Pilgangoora, North-West Terr., Australia	XXV, mauvette	—	1.595	1.599	2V=44½°	This paper

The refractive indices of rose muscovites are consistently lower, by 0.01–0.02, than the average values for 250 specimens of other pegmatitic muscovites, and the 2V values for rose muscovite are higher than those of the other pegmatitic muscovites. The purple muscovite from Pilar, N.M. (Schaller and Henderson, 1926) is optically not a rose muscovite, as here defined. Its color is much deeper, with strong pleochroism, the indices of refraction are unusually high: $\alpha = 1.565$, $\beta = 1.597$, $\gamma = 1.602$, $2E = 77°$. Jakob (1925) also has described an allegedly pale rose muscovite, but examination of a specimen of his analyzed material shows it to be a typical "ruby" muscovite, both in color and indices.

Structure

As demonstrated by Levinson (1952) there is a strong correlation in the muscovite-lepidolite series between Li_2O content and structure. Muscovite (0–3.3% Li_2O) is invariably two-layered monoclinic; lithian muscovite is two-layered monoclinic with consistent, small-scale departures from the normal muscovite structure; most micas of this series with 3.4–4.0% Li_2O are structurally transitional, consisting of both 2-layered and 6-layered polymorphs; lepidolites are 6-layered (4.0–5.1% Li_2O) or 1-layered monoclinic (>5.1% Li_2O). All of the rose muscovites studied have the two-layered monoclinic normal muscovite structure and normal cell dimensions.

Chemistry

Selected previously published analyses of rose muscovites are collected in Table 2. They show little variation in SiO_2 or Al_2O_3. Total iron is very low, normally less than 1%, and, where determined, MnO or Mn_2O_3 is present in small amounts. Li_2O is very low, usually less than 0.5%.

Five new analyses of rose muscovites are presented in Table 3. Again variations in SiO_2 and Al_2O_3 are not significant; Li_2O contents are similarly very low; total iron is very low, and MnO exceeds total iron in all but No. 1. Fluorine averages a little less than 1%. The formulae, which show the close relation to other muscovites, are:

1. $(K_{1.7}Na)_{1.9}(Mn_{0.02}, Mg, Fe^{2})_{0.03}(Al_{3.8}, Li_{0.02}Fe^{3}_{0.02}Ti)_{3.8}(Si_6, Al_2)_8O_{19.7}OH_4F_{0.3}$.
2. $(K_{1.7}, Na, Rb, Ca)_{2.0}Mn_{0.06}(Al_{3.6}, Li_{0.2}, Fe^{3})_{3.8}(Si_{5.9}, Al_{2.1})_8O_{19.6}OH_4F_{0.4}$.
3. $(K_{1.7}, Na, Ca)_{1.9}Mn_{0.04}(Al_{3.3}, Li_{0.3})_{3.6}(Si_{5.9}, Al_{2.1})_8O_{19.4}OH_{4.1}F_{0.5}$.
4. $(K_{1.8}, Na, Ca)_{2.2}Mn_{0.1}(Al_{3.4}, Li_{0.1})_{3.5}(Si_{5.9}, Al_{2.1})_8O_{19.9}OH_{3.1}F_{0.3}$.
5. $(K_{1.7}, Na, Rb, Cs)_{2.0}(Mn_{.02}, Mg_{.02})_{.04}(Al_{3.8}, Li_{0.3})_{4.1}(Si_6, Al_2)_8O_{19.9}OH_{3.7}F_{0.4}$.

The rose muscovites are low Fe, low Mn, normal Si muscovites, with consistent minor Li, F, Rb, and Cs. The rose muscovites, despite their color, cannot be grouped with alurgite, which is characterized by high SiO_2 content and is best classed as ferrian phengite. They also differ chemically from the red mica from Cajon Pass, Calif., so-called "alurgite,"

TABLE 2. SELECTED OLDER ANALYSES OF ROSE MUSCOVITES

	1	2	3	4	5	6	7	8	9	10	11	12	13	14
SiO_2	47.02	—	46.28	46.81	46.80	44.80	—	45.63	45.58	45.56	46.01	47.64	45.77	45.22
TiO_2	—	—	—	0.01	0.01	—	—	tr.	0.00	0.00	—	0.00	0.10	n.d.
Al_2O_3	36.83	—	36.86	36.09	35.84	37.72	—	37.42	37.45	29.53	35.64	34.22	37.02	37.46
Fe_2O_3	0.51	—	0.97	0.25	0.24	0.67	—	tr.	0.16	0.12	0.13	0.10	0.75	tr.
FeO		—				n.d.	—	—	—	0.10	—	—	—	0.56
MnO	1.05	—	tr.	n.d.	—	—	—	.06	0.15	2.32	0.09	0.05	—	0.71
Mn_2O_3	—	—	—	—	—	0.21	—	—	—	—	—	—	—	—
MgO	0.26	—	—	0.62	0.56	—	—	0.00	0.13	0.29	0.04	0.28	0.34	0.38
CaO	—	—	—	0.29	0.29	—	—	0.00	0.28	0.15	1.12	0.00	0.12	0.10
Li_2O	0.30	0.64	0.26	n.d.	—	tr.	0.38	0.20	0.13	1.80	0.69	1.10	0.26	0.32
Na_2O	—	0.99	1.41	0.68	0.60	—	0.91	1.43	0.93	2.65	1.88	0.47	1.17	1.19
K_2O	9.80	9.08	10.63	10.24	10.08	10.66	10.07	9.95	10.90	11.01	8.19	10.40	9.28	10.71
Rb_2O	—	—	—	n.d.	—	—	0.82	—	—	tr.	1.20	0.35	—	—
Cs_2O	—	—	—	n.d.	—	—	0.09	—	—	0.33	0.20	0.75	—	—
H_2O-	—	—	—	0.42	0.64	4.52	—	—	—	3.12	0.08	0.10	0.86	—
H_2O+	3.90	—	4.83	5.00	5.05		—	4.43	3.16		4.65	3.62	1.39	3.68
F	0.52	1.89	0.377	n.d.	-	0.20	—	0.77	0.97	3.45	0.54	1.21	0.44	0.73
	100.19	—	101.257	100.41	100.11	100.18	—	99.89	99.84	101.43	100.46	100.29	99.18	101.06
O=F	0.22	—	0.158	—	—	.08	—	0.32	0.41	1.45	0.23	0.51	0.18	.31
Total	99.97	—	101.099	100.41	100.11	100.10	—	99.57	99.43	99.98	100.23	99.78	99.00	100.75

1. Goshen, Mass. Rammelsberg, 1875.
2. Goshen, Mass. Mallet, 1857.
3. Haddam Neck, Conn. Bowman, 1903. P_2O_5– 0.09. "The MnO is sufficient to give the fused carbonates a pale bluish-green tinge, . . ." Bowman, 1902.
4., 5. Rutherford, Va. Glass, 1935. Iron determined as Fe_2O_3 and recalculated as FeO.
6. Harding, N. M. Schaller and Henderson, 1926.
7. Harding, N. M. Stevens and Schaller, 1942. Another determination by Ahrens (1948) gave Rb_2O—0.17, Tl_2O—0.015.
8. Mesa Grande, Calif. Clarke, 1915.
9., 10. Pointe du Bois, Manitoba. Ellsworth, 1932.
11., 12. Varuträsk, Sweden, Berggren, 1940.
13. Suizawa, Japan. Iimori and Yoshimura, 1929. (Cu, Pb)O—1.65, rare earths—0.03. Note low H_2O+.
14. Londonderry, Western Australia. Simpson, 1927.

TABLE 3. NEW ANALYSES OF ROSE MUSCOVITE

	1	2	3	4	5
SiO_2	45.00	45.03	45.60	45.54	45.24
TiO_2	0.02	0.02	0.03	0.07	0.01
Al_2O_3	37.41	36.33	35.96	36.36	36.85
Fe_2O_3	0.12	0.14	0.10	0.25	0.09
FeO	0.05	0.02	0.02	0.02	0.02
MnO	0.15	0.41	0.37	0.80	0.12
MgO	0.06	0.05	0.05	0.07	0.08
Li_2O	0.02	0.41	0.63	0.06	0.49
CaO	0.03	0.09	0.08	0.09	0.00
Na_2O	0.63	0.69	0.59	0.57	0.64
K_2O	10.50	10.50	10.52	10.76	10.08
Cs_2O	n.d.*	0.06	n.d.*	n.d.*	0.20
Rb_2O	n.d.*	0.79	n.d.*	n.d.*	0.93
H_2O+	4.48	4.56	4.81	4.35	4.12
H_2O-	0.68	0.79	0.44	0.55	0.46
F	0.78	1.01	1.31	0.62	0.91
	99.93	100.45	100.51	100.11	100.24
$0=F$	0.33	0.42	0.55	0.26	0.38
Total	99.60	100.03	99.96	99.85	99.86

Analyst: Eileen H. Oslund, Univ. Minnesota, Rock Anal. Lab.

* n.d., not determined: Cs and Rb weighed and calculated as K.

Li_2O, Cs_2O, Rb_2O by flame spectrophotometer, R. B. Ellestad, *analyst.*

1. Birkeland 2, Iveland, Norway.
2. White Spar No. 1, Gunnison Co., Colo.
3. Brown Derby No. 1, Gunnison Co., Colo.
4. Apache, Petaca, N. M.
5. Pittlite, Rociada, N. M.

of Webb (1939) and the reddish mica from Pilar, N. M., of Schaller and Henderson (1926), which are ferrian muscovites. Neither alurgite nor ferrian muscovite contain lithium, whereas the rose muscovites always contain small amounts of that element.

Minor Elements

The various minor elements recorded spectrographically in rose muscovites are listed in Table 4. Included also are the contents of Fe_2O_3 and MnO, which are in general agreement with the results of wet chemical analysis on other samples from the same localities. Ahrens (1948) has demonstrated that the Harding mica is unique in its very low Rb-Tl ratio, $Rb_2O/Tl_2O=12$. His muscovite with the next lowest ratio has $Rb_2O/Tl_2O=30$. Glass (1935) for No. 2, Table 2 determined the absence of BeO and the presence of B and Sn. Table 4 shows that the rose muscovites consistently contain significant quantities of Rb, and most of them also have small amounts of Cs. Ga and Sn are usually present, as is Ba in very minute amounts. Cr, V, Sr, Si and Co are very rarely present or are absent entirely. The absence of Sc is in general accord with the results of Oftedal (1943).

TABLE 4. MINOR ELEMENTS IN ROSE MUSCOVITES

	1	2	3	4	5	6	7	8
Fe_2O_3	0.12	0.12	0.15	0.08	0.19	0.04	0.09	0.12
MnO	0.17	0.38	0.25	0.45	0.95	0.10	0.22	0.15
Cr_2O_3	—	0.0001	0.0001	0.0001	—	—	—	—
BaO	0.0003	0.0004	0.0003	0.0005	0.002	—	0.0003	0.001
Rb_2O	0.29	0.41	1.0	1.8	0.26	0.17	0.62	0.46
Cs_2O	—	0.17	0.08	0.17	0.12	—	0.28	0.07
SrO	—	—	—	0.003	—	—	—	—
SnO_2	0.002	0.04	0.03	0.07	0.004	0.005	—	0.05
Ga_2O_3	0.008	0.019	0.037	0.05	0.035	0.009	0.014	0.021
Sc_2O_3	—	—	—	—	—	—	-	—
V_2O_5	—	—	—	—	—	—	0.0004	—
Co_2O_3	—	—	—	—	—	—	—	—

Spectrochemical determinations by Charles E. Harvey.

1. Birkeland 2, Iveland, Norway.
2. Höydalen, Telemark, Norway.
3. White Spar No. 2, Colorado.
4. Brown Derby No. 1, Colorado.
5. Apache, Petaca, New Mexico.
6. Pittlite, Rociada, New Mexico.
7. Harding, New Mexico.
8. Bolton, Massachusetts.

CAUSE OF COLOR

Several suggestions have been advanced in previous studies regarding the cause of color of rose muscovites. Iimori and Yoshimura (1929), who report 1.65% of (Cu, Pb)O in their analysis, believed that the color results from the presence of the Cu in the colloidal state. Spectroscopically they also detected Sc, Y, La, Eu, Gd, Dy, and Er, but regarded the quantity of Er and Eu too low to cause the pigmentation. Micke (1950) postulates a rhythmic variation in F to account for pale violet zones in muscovite, but offers no supporting evidence. Webb (1939) thought Ti caused the red color of the California "alurgite," but Meixner (1939) pointed out that other micas of similar general composition and of similar Ti contents are not red. Co was suggested as a possible coloring agent, but Table 4 shows that the Co content of rose muscovites is nil.

Because of the color similarity to some lepidolites, a few investigations have attempted to relate the rose color of these muscovites with Li content. Ellis (1950) for example, in describing the replacement of feldspar in the Main Tantalite Dike at Strelley by fine-grained pale purple chalcedony that grades into cryptocrystalline rose muscovite suggests that the color of both is due to Li. There is, however, no correlation even in lepidolites between color and Li content. High-lithium lepidolites may be purple, gray, or even white.

That the rose muscovite color depends on the presence of Mn was advanced early and has since been reemphasized by several workers. Mallet (1857) stated with regard to the Goshen mica, ". . . and there can I think be little doubt that both are essentially potash-micas of the species muscovite, the rose-colored being probably tinged by oxyd of manganese." Likewise Bowman (1902) reiterated that, "The MnO . . . is probably the cause of the pink colour of the mineral, but it does not appear to be present in weighable amounts." Schaller and Henderson (1926, p. 11) state, "In chemical composition there is very little variation in these micas from that of normal muscovite. Small quantities of iron and manganese are present as inherent parts of the mineral, probably replacing alumina (sic), and their combined color effect produces the abnormal color and pleochroism of the mineral." There exist today numerous analyses of non-rose or non-purple muscovites that contain small amounts of MnO as well as amounts of Fe_2O_3 considerably above that reported by Schaller and Henderson (1926) in the Harding mica, which has $Fe_2O_3 = 0.67\%$. It seems unlikely, therefore, that the combined effect of small amounts of these ions brings about an abnormal coloration that cannot be produced by larger quantities of the same elements.

As has been pointed out by Shibata (1952) the pigmenting agents in muscovite are Fe^2, Fe^3 and Mn; Ti is normally negligible in muscovites. In an ordinary mica analysis the valence of Mn cannot readily be determined, and any Mn present is usually reported as MnO. If FeO is present in the muscovite it is to be presumed that Mn is present only as Mn^2, for Fe is more easily oxidized than Mn:

	E_0 (in volts)
$Fe^{2+} \rightarrow Fe^{3+} + e$	0.77
$Mn^{2+} \rightarrow Mn^{3} + e$	1.51

In most muscovites FeO, or $FeO + Fe_2O_3$, is greatly in excess of MnO, and any coloration effect that Mn^2 could contribute is masked by the effects of the Fe^2 or Fe^3 ions, for compounds of bivalent Mn are only faintly colored. However in muscovite in which Fe^2 is negligible and only small amounts of Fe^3 are present, the Mn can and may well exist as Mn^3, which is a strong chromophore. In the newly analyzed rose muscovites of Tables 3 and 4, FeO is nearly absent and $MnO > Fe_2O_3 + FeO$ in all micas but No. 1, in which $FeO+Fe_2O_3:MnO$ is$\cong$1:1. Micas 2–5 are distinctly rose colored, but No. 1, from Iveland, Norway, is of an exceedingly pale rose tint, and flakes in some parts of the aggregate are completely colorless or even a very faint green. The rose muscovites owe their delicate and unusual color, therefore, not to the mere presence of either Fe or Mn, nor even to their combined presence, but to the essential absence of Fe^2 and the equality or predominance of Mn^3 with respect to Fe^3.

This conclusion is substantiated by Jacobson and Webb (1946) who found that in their zoned mica books the green part had a higher Fe/Mn ratio than the rose part. Further support is offered, for example, by two analyses of muscovites by Ellsworth (1932, pp. 266–267). The two curved micas occur together in the Annie pegmatite. The lilac (No. 10, Table 2) contrasts with the gray:

	Lilac	*Gray*
Fe_2O_3	0.12	2.20
FeO	0.10	2.57
MnO	2.32	0.90
Li_2O	1.80	0.90

In lepidolites, the color variations also are related to fluctuations in the Fe/Mn ratio. The role of Mn in the coloration of lepidolite has been stressed, for example, by Hintze, 1897 and by Doelter, 1917. As stated by Shibata (1952 p. 160), "Lepidolite is pink or violet, which is due to the color of manganese. Lepidolite is sometimes colorless when it is poor in MnO. Lithium is not a color agent. . . ." Those lepidolites that are colorless or of very pale colors usually have Fe:Mn$\cong$1:1; those that are gray normally have Fe>Mn, whereas the purple lepidolites contain more Mn than Fe.

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SYNTHESIS OF GRAPHITE AT ROOM TEMPERATURES*

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Abstract

Available thermodynamic data indicate that the cubic modification of carbon is the stable form at room temperatures and pressures above 15,000 atmospheres. Elemental carbon was formed under pressures as high as 35,000 atmospheres by the reaction: mercuric carbide→mercury+carbon. The carbon was identified as graphite by *x*-ray powder patterns.

The experimental procedure was to enclose the mercuric carbide powder in sealed copper cylinders which were compressed by hydraulically operated tungsten carbide pistons in a hollow steel cylinder. The rate of decomposition was unpredictable and more often than not proceeded explosively.

Introduction

The extensive literature on the synthesis of crystalline carbon is exclusively devoted to experiments carried on at elevated temperatures. In most of the methods described the temperatures are so high that it would be impossible to sustain high pressures along with the temperatures for any appreciable length of time because the confining vessel would fail at the temperatures used. Some experimentation, such as Hannay's (1) discredited attempts to produce the diamond, has been done at only moderately elevated temperatures. It should also be noted that the temperature of formation of carbon black is comparatively low but the product does not give *x*-ray diffraction patterns that would warrant its being described as distinctly crystalline.

The available thermo-dynamic data (2) indicate that graphite is the stable form of carbon at all temperatures if the pressure is below 15,000 atmospheres, and that diamond is the stable form at higher pressures, if the temperatures are not greatly above room temperature. The transition boundary at moderate temperatures falls not only well within the limits of convenient laboratory experimentation but also within the range of industrial processes. On the basis of these data Bridgman (3) has unsuccessfully attempted to convert small flakes of graphite to diamond. The failure of an attempt to convert one allotropic form to another does not vitiate the thermo-dynamic data because of the strong tendency of one form to remain in a metastable condition after the transition boundary has been passed. The adaption of such a transformation to practical uses is greatly limited by the particle size of the product if, as appears to be true in this instance, conditions are unfavorable for crystal growth by accretion.

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* Contribution from the Department of Mineralogy and Petrography, University of Michigan, No. 170.

The objective of this investigation was to discover a reaction, one product of which would be elemental carbon whose rate of formation could be controlled and which would proceed under conditions that would permit crystal growth by accretion. Without describing unsuccessful attempts it can be stated that the reaction: mercuric carbide→carbon +mercury, occasionally fulfills all the above conditions, yielding graphite. However, in most instances the decomposition is either only slight or proceeds explosively. The reaction is well adapted to high pressure experimentation because the carbide is a dry white powder and the liquid product mercury forms a solid amalgam with excess copper thus eliminating many of the difficulties of sealing off gases and liquids under high pressure.

EXPERIMENTAL PROCEDURE

Figure 1*a* is a cross section of the pressure assembly which is actuated by a hydraulic ram. The thrust of the ram is transmitted through the upper solid steel cylinder which drives a tungsten carbide piston $\frac{1}{4}$ inch

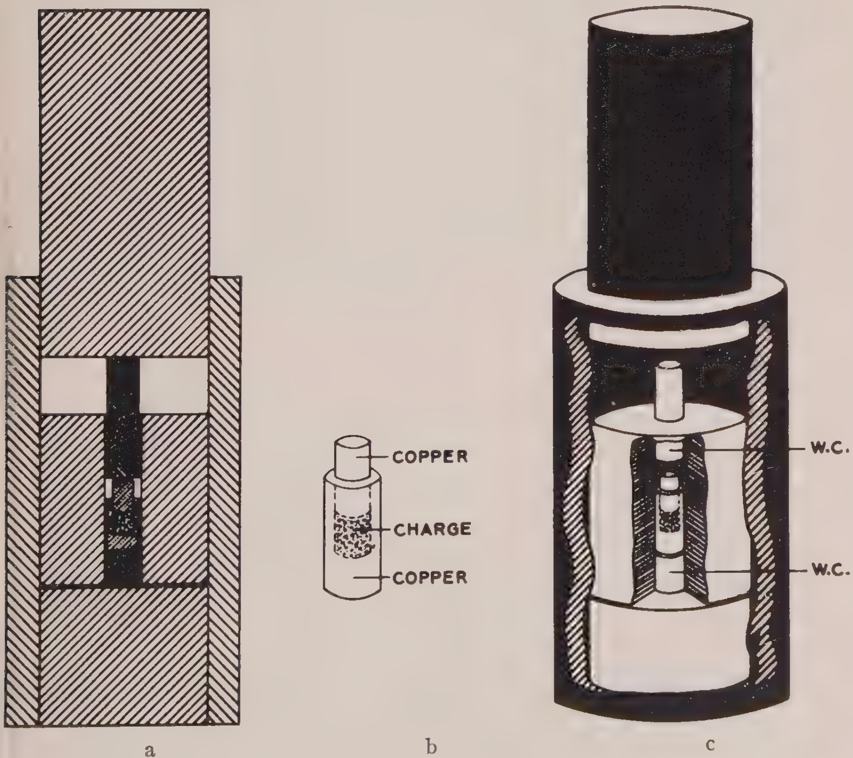

FIG. 1. Pressure assembly.

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in diameter into the pressure cylinder. A small tungsten carbide plug closes the other end of the pressure chamber and supports the copper jacketed sample, Fig. 1*b*. The cylinder rests on a solid steel cylinder of the same diameter and all is enclosed in a steel jacket that serves to keep the component parts in alignment and also gives protection from flying chips if the protruding end of the $\frac{1}{4}$ inch piston fails. The sample, approximately one-quarter gram in weight, is placed in a $\frac{1}{4}'' \times \frac{3}{8}''$ drilled copper cylinder, Fig. 1*b*, and a copper plug is inserted in the open end. Under compression the plug cold welds to the wall of the copper cylinder when the yield point of copper is exceeded. Figure 1*c* is a cut-away sketch of the assembly ready to be placed under the hydraulic ram. The several cylindrical surfaces are ground to a smooth sliding fit.

An electrically driven oil pump of 10,000 psi capacity powers the ram through a bypass control valve which can be regulated between 500–6,000 psi. The cross-sectional area of the ram piston is slightly over five square inches which when driving the $\frac{1}{4}''$ diameter tungsten carbide piston gives a hydraulic multiplication of 103. When pressure on this piston exceeds the yield point of copper, the jacket enclosing the sample flows plastically and transmits the pressure hydrostatically to the sample. Tissue thin sheets of copper are extruded 2–3 mm. above and below the tungsten carbide piston between it and the walls of the pressure chamber. In several instances when the steel of the pressure chamber failed the copper flowed into the cracks developed, Fig. 2.

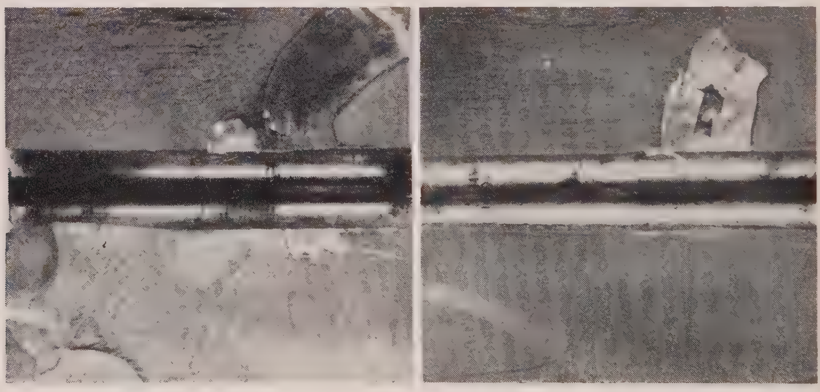

FIG. 2. Halves of steel pressure cylinders that cracked showing tissue thin sheets of copper that were extruded into the cracks.

At the conclusion of an experimental run, the copper case with the enclosed sample is forced out of the pressure cylinder with a hand operated hydraulic press. It is then cut in half by rolling under a knife edge, the sample removed, digested in nitric acid or a solution of potassium cyanide and washed. Graphite when produced was identified by *x*-ray powder patterns. The reaction is highly erratic and unpredictable. Often decomposition proceeds only far enough to darken the sample even after several days under pressure. After dissolving these samples there remains a flocculent black residue too small to be positively identified. Several samples were lost by spontaneous explosions while cutting through the case or while forcibly removing the compacted sample with a needle point. The greatest source of failure which accounts for the loss of three out of four samples is the explosive decomposition while bringing the pressure up to the desired point. This is easily recognized by the sharp sound and the shock which is transmitted to the pressure gauge. If the pressure is sufficiently high at the time of explosive decomposition the pressure cylinder ruptures and a fine black soot is driven into the available openings. The carbon produced by explosive decomposition is too finely divided to give an *x*-ray powder pattern.

On opening the copper cases the first impression, in those instances when graphite was produced, was that it had the high luster of natural graphite. Under the microscope it was apparent that the luster was due to the multitude of minute spherical globules of mercury on the surface. Nitric acid or solutions of potassium cyanide were used to dissolve the mercury and the sample then broke up into irregular fragments with a dull luster and a somewhat porous appearance. These fragments crushed readily but offered more resistance than was anticipated. This first impression of grittiness was lost on crushing. *X*-ray powder patterns gave sharp distinct lines with no evidence of preferred orientation which is so characteristic of graphite. No attempt was made to determine whether both forms of graphite were present.

DISCUSSION

Several methods of preparing mercuric carbide were used, all based on passing acetylene gas into solutions of mercuric salts. Two compounds, HgC_2 and $HgC_2 \cdot \frac{1}{2}\ H_2O$ have been previously reported. The hydrate would have 3.4% H_2O but its determination is fraught with difficulty because of the tendency of freshly prepared samples to decompose explosively around 100° C. All water however is removed in a vacuum desiccator. One near serious accident occurred in attempting to dry samples of the freshly prepared carbide in an oven at 110° C.

In the early stages of the investigation it was thought that particle size exercised a control over the tendency to explode because rapid precipitation yielded material that exploded readily with increased temperature or pressure. *X*-ray powder patterns from these samples gave broad diffuse lines indicative of minute crystals. Samples prepared by the slow precipation from dilute solutions showed less tendency to explode and gave sharp lines in x-ray patterns. Mercuric carbide precipitated slowly from a dilute $KHgI_3$ solution gave the most satisfactory results and it was from such samples that graphite was produced. However, working under the assumption that particle size controlled the tendency towards spontaneous decomposition, little progress was made toward the preparation of samples from which similar results could be obtained under apparently identical conditions. The experiments were conducted between the temperature range of solid CO_2 and 125° C. and pressures up to 30,000 atmospheres. The results were very erratic and temperature had no apparent influence upon the rate of reaction.

In September 1952 samples which had aged over the summer were found to be stable in the range of pressure and temperature of the previous experiments. They could also be analyzed by distilling over the mercury in a vacuum at 200° C. and after 6 days at 35,000 atmospheres at room temperature decomposition had only proceeded far enough to darken the sample. A reexamination of previous work showed that an hypothesis of stabilization by aging could account for the erratic behavior that had previously been encountered.

Such an hypothesis can appear plausible by assuming that in the rapid precipitation of such a highly insoluble compound many centers of disorder would develop within the crystals because of the rapidity with which the atoms must fall in place. In such a structure with many centers of disorder, each unstable with respect to the whole, a rise in temperature or pressure could initiate decomposition within one which would activate adjacent centers until enough energy was released to detonate the sample. If during the process of aging, time allows the disorder to partially heal and to reach a relative degree of equilibrium, the sample itself may become stable. In experimental procedures in which negative results are the rule and success comes occasionally by accident one must of necessity have recourse to any hypothesis which indicates avenues of further experimentation. In this instance it appears that in well aged samples a controlled rate of decomposition may be attained if they are subjected to somewhat higher temperatures, higher pressure or both.

In Fig. 3, adapted after Rossini (4), the interval between the x's shows the range in which graphite was produced. This is well within the region where diamond should be the stable modification but no conclusions are warranted as to whether these results indicate that the diagram is inaccurate or whether other factors may exercise control over the modification that is formed. It has been suggested that the decomposition of a compound in which the carbon is tetrahedrally bonded in a manner analogous to that of the diamond might favor the formation of

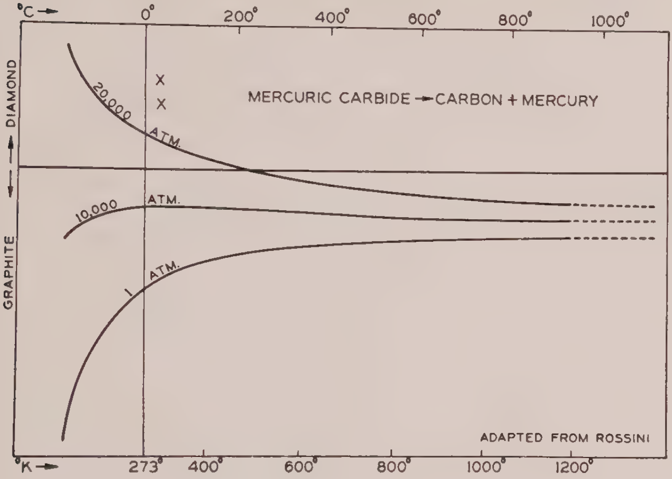

FIG. 3. Diamond-graphite phase diagram.

the cubic modification. The structure of mercuric carbide is similar to that of calcium carbide and the dissimilarity of the structure to that of diamond may militate against its success. However, carbon is appreciably soluble in mercury (5), especially at higher temperatures, and hence this reaction may afford the possibility of crystal growth by accretion.

The few positive results of the investigation bear little relationship to the time and effort expended. A serious handicap to one considering the synthesis of the diamond is the meager information available about the negative results of the innumerable attempts that have been made in the past few decades. Much of this past work has been at extremely high temperatures and without doubt there has been a great deal of duplication of effort. The evidence afforded by the natural occurrence of diamond indicates that the temperatures of formation were not greatly in excess of 1000° C. The existing thermodynamic data support this conclusion.

This investigation has been carried on with the financial assistance of the Office of Naval Research. Much of the chemical work and all of the *x*-ray studies have been done by Mr. Richard Mitchell and Mr. A. A. Giardini.

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A NEW HEXAGONAL POLYMORPH OF SILICON CARBIDE, 19H*†

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ABSTRACT

Morphological and structural details of silicon carbide type 19*H* are given. The crystal has a structure represented by the zigzag sequence 22232323. The space group is *P3m*. The unit cell dimensions are $a_0=3.073$ kX and $c_0=47.75_3$ kX; $Z=19$.

Weissenberg photographs of silicon carbide modification 19*H* were first made in 1950. These were of very poor quality and only indicated that the crystal under investigation was an intergrowth of 6*H*, 15*R* and some other type which was not clear. The crystal was laid aside until 1952 when an intensive reinvestigation of a number of questionable crystals was undertaken using the Buerger precession and Laue methods. Excellent Buerger precession films were obtained of this modification as well as other previously unknown rhombohedral types consisting of many layers, e.g., 141*R*, 168*R*, 192*R*. Some of these will be reported in a forthcoming paper.

The existence of modification 19*H* was predicted by Ramsdell and Kohn (1952). They also predicted for its structure the zigzag sequence 22232323. It is assumed that the only numbers found in silicon carbide zigzag sequences are 2, 3 and 4 (Ramsdell and Kohn, 1952). No sequence involving the number 4 will complete a 19 layer hexagonal cell. The only possible way to complete this cell using 2 and 3 is the arrangement 22232323. The calculations for this sequence gave satisfactory agreement with the observed intensities of the films. Table 1 compares the calculated and observed intensities for this sequence. The observed intensities are an average of the results of a number of observers. The presence of intergrowths of 6*H* and 15*R* gave reflections which made the evaluation of the intensities of the 19*H* spots impossible in the case of 10·6, 10·3, and 10·13. The presence of these "foreign" spots was also considered in evaluating the intensities of the other reflections listed in the table.

The 19*H* zigzag sequence 22232323 includes the 15*R* sequence 232323. As a consequence there should be a marked similarity in their reflections. This is most easily observed in the comparison of the intensities of the 10·*l* reflections. On Weissenberg or precession films the rows of 10·*l* reflections of all silicon carbide types coincide. Moreover, the *range* from 10·0 to 10·19 for 19*H* coincides exactly with the *range* 10·0 to 10·15 in

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† This investigation was made possible by assistance from the Office of Naval Research.

TABLE 1. COMPARISON OF OBSERVED AND CALCULATED INTENSITIES FOR SOME OF THE REFLECTIONS OF TYPE 19*H*

(10·*l*)	$I_{calc.}$	$I_{obs.}$
10·0	1.7	*m*
1	12.6	*s*
2	0.5	*vw*
3	0.0	—
4	0.2	*vvw*
5	100.0	*vvs*
6	1.4	—
7	0.8	*w*
8	2.4	*m*
9	88.2	*vvs*
10	1.6	*vw*
11	0.9	*vw*
12	3.3	*m*
13	12.2	*s*
14	1.4	*vw*
15	1.0	*vw*
16	6.8	*ms*
17	3.3	*m*
18	0.8	*vw*
19	0.5	*vvw*
10·$\bar{1}$	2.4	*m*
$\bar{2}$	14.5	*s*
$\bar{3}$	21.8	*s*
$\bar{4}$	3.8	*ms*
$\bar{5}$	4.0	*ms*
$\bar{6}$	39.8	*s*
$\bar{7}$	19.9	*s*
$\bar{8}$	2.1	*w*
$\bar{9}$	2.0	*w*
$\overline{10}$	72.4	*vs*
$\overline{11}$	1.3	*vw*
$\overline{12}$	0.3	*vvw*
$\overline{13}$	0.3	—
$\overline{14}$	28.6	*s*
$\overline{15}$	0.0	*a*
$\overline{16}$	0.0	*a*
$\overline{17}$	0.3	*vvw*
$\overline{18}$	3.7	*ms*
$\overline{19}$	0.3	*vvw*

15*R*. However, because of the rhombohedral extinctions of 15*R*, the only 10·*l* reflections are those with $l=1$, 4, 7, etc. and $\bar{2}$, $\bar{5}$, $\bar{8}$, etc. The 19*H* reflections which do not approximately coincide with 15*R* positions are either weak or absent. The 19*H* reflections which coincide most closely with 15*R* reflection positions are of stronger intensity, and, of these, those near very intense 15*R* reflection positions have the greatest intensity. For example, intensity calculations for 19*H* show $10\cdot 5$ to be the strongest reflection. It is nearest in position to $10\cdot 4$ for 15*R*, which likewise is the strongest 15*R* reflection. But the calculated intensity for $10\cdot \bar{5}$ of 19*H* is very low, and this corresponds to $10\cdot 4$, which is a missing reflection in the 15*R* lattice. Similar relationships are shown for other reflections in Table 2, in which the strongest 19*H* reflections are given.

TABLE 2. A COMPARISON OF THE INTENSITIES OF SOME 19*H* AND 15*R* REFLECTIONS

19*H*	$10\cdot 17$– $10\cdot 16$	$10\cdot 13$– $10\cdot 12$	$10\cdot 9$	$10\cdot 5$	$10\cdot 1$	$10\cdot\bar{2}$– $10\cdot\bar{3}$	$10\cdot\bar{6}$– $10\cdot\bar{7}$	$10\cdot\overline{10}$	$10\cdot\overline{14}$	$10\cdot\overline{18}$
order	9	8	2	1	7	6	4	3	5	10
15*R*	$10\cdot 13$	$10\cdot 10$	$10\cdot 7$	$10\cdot 4$	$10\cdot 1$	$10\cdot\bar{2}$	$10\cdot\bar{5}$	$10\cdot\bar{8}$	$10\cdot\overline{11}$	$10\cdot\overline{14}$
order	9	7	2	1	8	5	4	3	6	10

The 15*R* reflections which they approach are listed directly under them. The numbers under each of these indicate decreasing intensities. From these orders one can see the correlation of the intensities of the 19*H* reflections with the corresponding 15*R* reflections. In a few cases where two 19*H* reflections are both close to a 15*R* reflection position, the stronger of these was used in determining the order of intensity.

This structure has the space group $P3m$,* as in the 10*H* polymorph. The unit cell dimensions are $a_0=3.073$ kX,† $c_0=47.75_3$ kX, and $Z=19$. The zigzag sequence 22232323 results in the following atomic positions:

19 Si at $000, 002z, 004z, 008z, 0011z, 0015z, 0017z,$
$\frac{1}{3}\,\frac{2}{3}\,1z,\ \frac{1}{3}\,\frac{2}{3}\,5z,\ \frac{1}{3}\,\frac{2}{3}\,7z,\ \frac{1}{3}\,\frac{2}{3}\,9z,$
$\frac{1}{3}\,\frac{2}{3}\,13z,\ \frac{1}{3}\,\frac{2}{3}\,16z,\ \frac{2}{3}\,\frac{1}{3}\,3z,\ \frac{2}{3}\,\frac{1}{3}\,6z,$
$\frac{2}{3}\,\frac{1}{3}\,10z,\ \frac{2}{3}\,\frac{1}{3}\,12z,\ \frac{2}{3}\,\frac{1}{3}\,14z,\ \frac{2}{3}\,\frac{1}{3}\,18z.$
19 C at $00p, 002z+p, 004z+p, 008\,z+p, 00\,11z+p, 00\,15z+p,$
$00\,17z+p,\ \frac{1}{3}\,\frac{2}{3}\,1z+p,\ \frac{1}{3}\,\frac{2}{3}\,5z+p,\ \frac{1}{3}\,\frac{2}{3}\,7z+p,$
$\frac{1}{3}\,\frac{2}{3}\,9z+p,\ \frac{1}{3}\,\frac{2}{3}\,13z+p,\ \frac{1}{3}\,\frac{2}{3}\,16z+p,$
$\frac{2}{3}\,\frac{1}{3}\,3z+p,\ \frac{2}{3}\,\frac{1}{3}\,6\,z+p,\ \frac{2}{3}\,\frac{1}{3}\,10z+p,$
$\frac{2}{3}\,\frac{1}{3}\,12z+p,\ \frac{2}{3}\,\frac{1}{3}\,14z+p,\ \frac{2}{3}\,\frac{1}{3}\,18z+p.$
$z=1/19,\ p=3/76$

The crystal is dark blue in color. It is roughly 1 mm. in diameter and

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* The hexagonal primitive lattice is designated as *P*, following the new *International Tables for X-ray Crystallography*.

† These dimensions are given in kX units to agree with the previously published values for the various SiC types.

TABLE 3. MORPHOLOGICAL DATA

Form	No. times observed	Quality	Observed	Calculated*
00·1	2	AC	0°00′	0°00′
10·0	3	DE	90°00′	90°00′
10·1	1	D	86°40′	86°49′
10·2	2	E	83°36′–83°50′	83°38′
10·3	2	CD	80°16′–80°25′	80°31′
10·4	1	D	77°34′	77°26′
10·5	2	CE	74°28′–74°31′	74°26′
10·6	1	E	71°26′	71°31′
10·7	0			68°41′
10·8	1	E	65°40′	65°58′
10·9	1	D	63°11′	63°22′
10·10	2	D	60°51′–60°54′	60°52′
10·11	0			58°29′
10·12	0			56°14′
10·13	3	DE	54°00′–54°34′	54°05′
10·14	0			52°02′
10·15	0			50°01′
10·16	0			48°16′
10·17	0			46°33′
10·18	0			44°54′
10·19	0			43°21′

* Calculated from the theoretical axial ratio for a 19 layer cell.

0.25 mm. thick. Table 3 gives the morphological data for those 19*H* faces measured on the crystal. The faces, which are confined to three pyramid zones, were in nearly every instance of very poor quality. No attempt was made to determine by etching which is the upper and which is the lower pinacoid (Thibault, 1944) and, thus, no distinction is made between the measured 10·l and 10·$\bar{l}$ faces.

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APPLICATION OF THE LAUE PHOTOGRAPH TO THE STUDY OF POLYTYPISM AND SYNTAXIC COALESCENCE IN SILICON CARBIDE*†

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Abstract

The purpose of this paper is to give a summary of the relationships between Laue photographs of various polytypic forms of silicon carbide. A knowledge of these relationships makes this method readily applicable to the (*a*) discovery of new polymorphs, (*b*) determination of syntaxic intergrowths and coalescence and, (*c*) identification of many-layered rhombohedral types.

The use of the optical goniometer in the study of polytypic substances as well as their syntaxic coalescence and intergrowths was greatly emphasized by Thibault (1944). The importance of the optical goniometer must still be emphasized, but recently, especially with the discovery of rhombohedral silicon carbide types with a large number of layers, its limitations are more clearly seen. In this laboratory, for example, crystals that showed blurred faces in the goniometer were usually discarded. Later it was learned that a great number of these are rhombohedral modifications of many layers These could only be identified with the use of the Laue methods which will be described. Now that this fact is known, it is possible to sort out, for *x*-ray investigation, those crystals that have possibilities of being new modifications. With our present knowledge of the polymorphs of silicon carbide, coalescence and syntaxic intergrowths are now more easily perceived and identified by *x*-ray methods than by the optical goniometer.

The Interpretation of Silicon Carbide Laue Photographs

Because type $6H$ is the most common polymorph of silicon carbide it will be used in the following discussion which is aimed at showing some geometrical relationships between the crystal faces, reciprocal lattices and Laue photographs of this substance.

In Fig. 1 lines AE, AF, AG, AH and AI represent the slopes of the $6H$ $10\cdot l$ faces as indicated. Lines AR_1, AR_2, AR_3, AR_4 and AR_5 are perpendicular to AE, AF, AG, AH and AI, respectively. The intersections of these AR_n lines with the gnomonic plane, represented by BR_1, give the gnomonic projection of the $10\cdot l$ faces (G_n). The intersections of the AR_n lines with DR_6 establish a reciprocal lattice network of these $10\cdot l$ faces. The distance between each of these is equal to DR_1. In this discussion we are concerned with the planes in the range D to R_6. Point R_6 was not established by the use of an external face since this face is not present on the crystals, but it must be used because we are considering a 6 layer form. The distance DR_6 represents the reciprocal distance of one layer of the structure. This single layer of $d = 2.513$ kX* is the basic unit in all silicon carbide forms. Line CM represents a flat Laue film. The L_n points represent the Laue spots of the $10 \cdot l$ reflections.

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† This investigation was made possible by assistance from the Office of Naval Research.

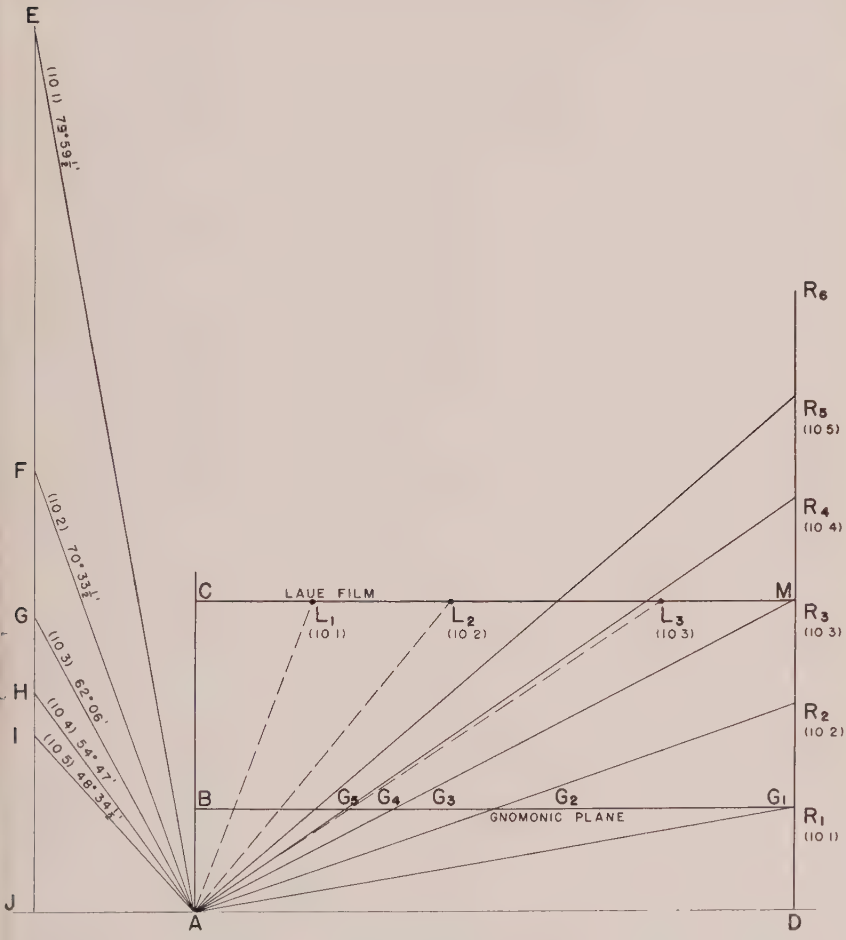

FIG. 1. Some geometrical relationships between the crystal faces, reciprocal lattice, gnomonic projection and Laue photograph of silicon carbide type 6*H*.

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* This dimension is given in kX units to agree with the previously published values for the various SiC types.

TABLE 1

n	DR_n (cm.)	CL_n (cm.)
1	.2	.21
2	.4	.42
3	.6	.64
4	.8	.85
5	1.0	1.07
6	1.2	1.29
7	1.4	1.51
8	1.6	1.73
9	1.8	1.95
10	2.0	2.19
11	2.2	2.42
12	2.4	2.66
13	2.6	2.91
14	2.8	3.16
15	3.0	3.42
16	3.2	3.68
17	3.4	3.96
18	3.6	4.24
19	3.8	4.53
20	4.0	4.84
21	4.2	5.15
22	4.4	5.49
23	4.6	5.83
24	4.8	6.19
25	5.0	6.57
26	5.2	6.97
27	5.4	7.40
28	5.6	7.84
29	5.8	8.32
30	6.0	8.83
31	6.2	9.37
32	6.4	9.95
33	6.6	10.58
34	6.8	11.25
35	7.0	11.99
36	7.2	12.78
37	7.4	13.66
38	7.6	14.62
39	7.8	15.70
40	8.0	16.88

The R_n points are related to the L_n points according to the equation $\angle CAL_n = 2\angle DAR_n$ (1). It is, therefore, easy to determine the positions of the $10 \cdot l$ spots on a Laue film if the R_n points are known.

It was stated above that the distance DR_6 is the reciprocal of the basic unit, 2.513 kX, of all silicon carbide forms. Because this is a reciprocal relationship it follows that this distance can be divided into as many spaces as there are layers in the structure built up of this basic unit. We saw above that this distance in the $6H$ modification was divided into 6 parts. In $8H$, $10H$ and $19H$ the distance DR_6 would be divided into 8, 10 and 19 spaces, and in these cases R_6 would become $10\cdot 8$, $10\cdot 10$ and $10\cdot 19$, respectively. Using equation (1) one can easily see from this discussion how the positions of the $10\cdot l$ spots (L_n) can be predicted on a Laue film from the known R_n positions of any form of silicon carbide. The rhombohedral forms can be predicted in the same manner as long as one remembers that the $10\cdot l$ reflections are missing when $-1+l\neq 3n$.

To facilitate the construction of Laue patterns of silicon carbide with a crystal to film distance of 6 cm., DR_6 was made equal to 12 cm. This distance was then arbitrarily divided into 60 R_n spaces and the corresponding L_n positions were determined by the formula $L_n=6 \tan 2\angle DAR_n$. It was only necessary to calculate the first 40 L_n positions because the flat Laue film is seldom large enough to register more than this. The relationships are summarized in Table 1. These CL_n distances are plotted in scale form in Fig. 2, where each line is one of the 40 L_n positions. Each of these positions on the Laue film represents a corresponding reciprocal lattice R_n position. Therefore, on the scale, these have been labelled in centimeters according to the DR_n spacings as shown in Table 1. With the use of this scale one only has to think about the DR_n spacings. In order to show by example how this scale is applied, the positions of the $10\cdot l$ Laue spots will be determined for the hexagonal form $19H$. Dividing 12 cm. (DR_6), by 19, the number of layers in the modification, gives the value 0.63 cm. This distance marked off on the scale (Fig. 2) gives the position of the first $19H$ $10\cdot l$ reflection, $10\cdot 1$. Multiples of this distance give the other $10\cdot l$ spots. The $10\cdot\bar{l}$ reflections are in the same positions on the opposite side of the origin. The procedure for the rhombohedral type $33R$ is the same. Twelve divided by 33 gives 0.363 cm. This distance on the scale gives $10\cdot 1$. Twice this distance in the opposite direction gives $10\cdot\bar{2}$. The other spots, both negative and positive, fall on equally spaced points at 3 times 0.363 cm.

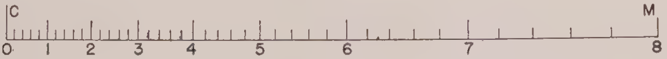

FIG. 2. Scale for the construction of silicon carbide Laue patterns with a crystal-to-film distance of 6 cm. (The scale has been reduced one-half.)

The writer has calculated the spot positions for numerous postulated as well as known forms of silicon carbide, and has arranged the results in two charts, one of the hexagonal modifications and one of the rhombohedral modifications. This latter chart is reproduced in Fig. 3.* If the chart is made to the same scale as the Laue crystal-to-film distance it is a relatively easy matter to slide the center of the film down the 0 line and find the corresponding spot positions, thus identifying the silicon carbide modification or modifications. One must, however, be aware of the fact that the 10·*l* spots are not the only ones present on the film. More often than not, especially in types consisting of a few layers, other spots appear in the midpoints between the 10·*l* spots, as well as at other odd positions. Only those at the midpositions give trouble because they frequently give the impression that the crystal form has twice as many

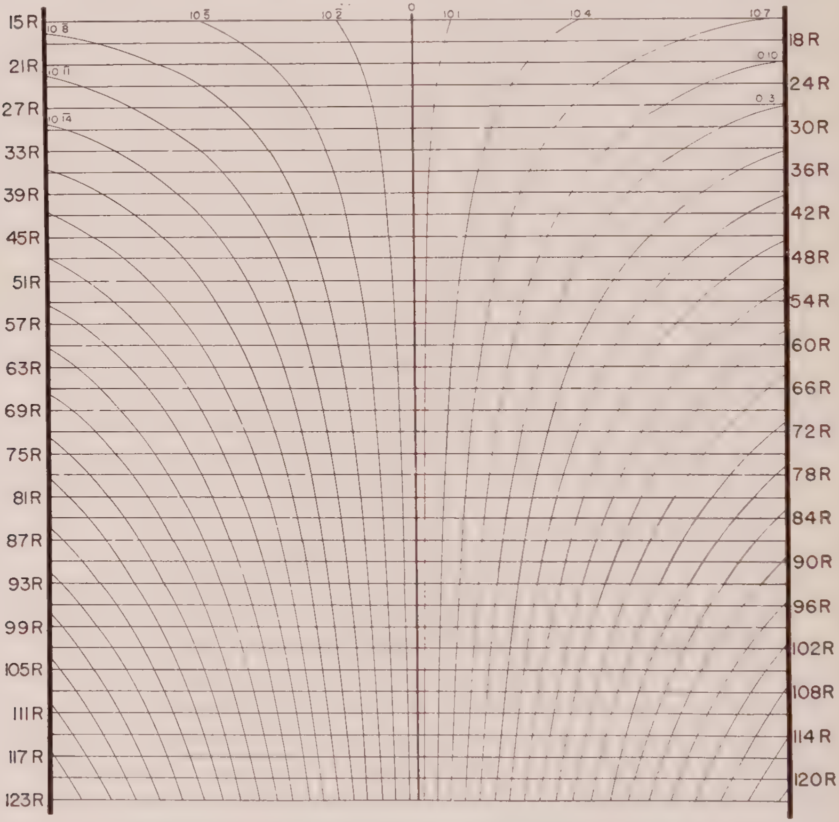

FIG. 3. Chart for the rapid identification of rhombohedral silicon carbide types from Laue films.

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* A number of the forms on this chart are theoretically impossible, but were included in order to give continuity to the chart.

layers to the unit cell than it actually has. This ambiguity is easily eliminated by further investigation with the rotating crystal methods. With a little experience one is not hindered greatly with the presence of these "unwanted spots." In a number of the many-layered rhombohedral modifications investigated in this laboratory, these have been much weaker than the $10 \cdot l$ spots or absent altogether.

Syntaxic coalescence is very common in silicon carbide. The effect of this property on the x-ray films is a superimposition of the patterns of each of the types. It is usually a very simple matter to differentiate between these superimposed patterns. This is true because the coalescence in nearly every case investigated consists of not more than three modifications and in these, two of the types were either $6H$, $15R$ or $4H$. Standard films of these should be available to make identification easier, but in case they are not available, one can determine the spots on the basis of the Laue pattern constructions described above. Actually, the coalescence of a common type in an unknown crystal is often helpful since the known spots are valuable reference points in determining the unknown form.

THE ADVANTAGES AND LIMITATIONS OF THE LAUE METHOD

The Laue method has a great advantage over most of the x-ray techniques when it comes to the many-layered rhombohedral forms, in that it resolves the spots much more than in these other conventional methods. Figure 4 shows the relative resolution of the $10 \cdot l$ reflections by three conventional x-ray methods, using CuK_α radiation. The crystal-to-film distance was 6 cm. for both the Laue and precession films, and the Weissenberg camera had a diameter of 5.73 cm. The over-all figure has been reduced in size. This greater resolving power of the Laue method has been used to advantage by other writers. Honjo, Miyake and Tomita (1950) were enabled, by observations taken from cylindrical Laue photographs, to identify their $594R$ silicon carbide. Ramsdell and Kohn (1952) also used Laue photographs to give sufficient resolution for the positive identity of their $84R$ polymorph. The recent positive identification of types $141R$, $168R$, and $192R$ at this laboratory was based upon Laue films.

Another important advantage of the Laue method is the speed at which one can prepare the crystal and take the photograph. Since the pictures are taken with the c axis parallel to the x-ray beam, it is a simple matter to orient them. Excellent basal pinacoids are nearly always present on crystals of this substance. The writer has observed many crystals which possess no other faces but the basal pinacoid. Using the Laue method these can be identified.

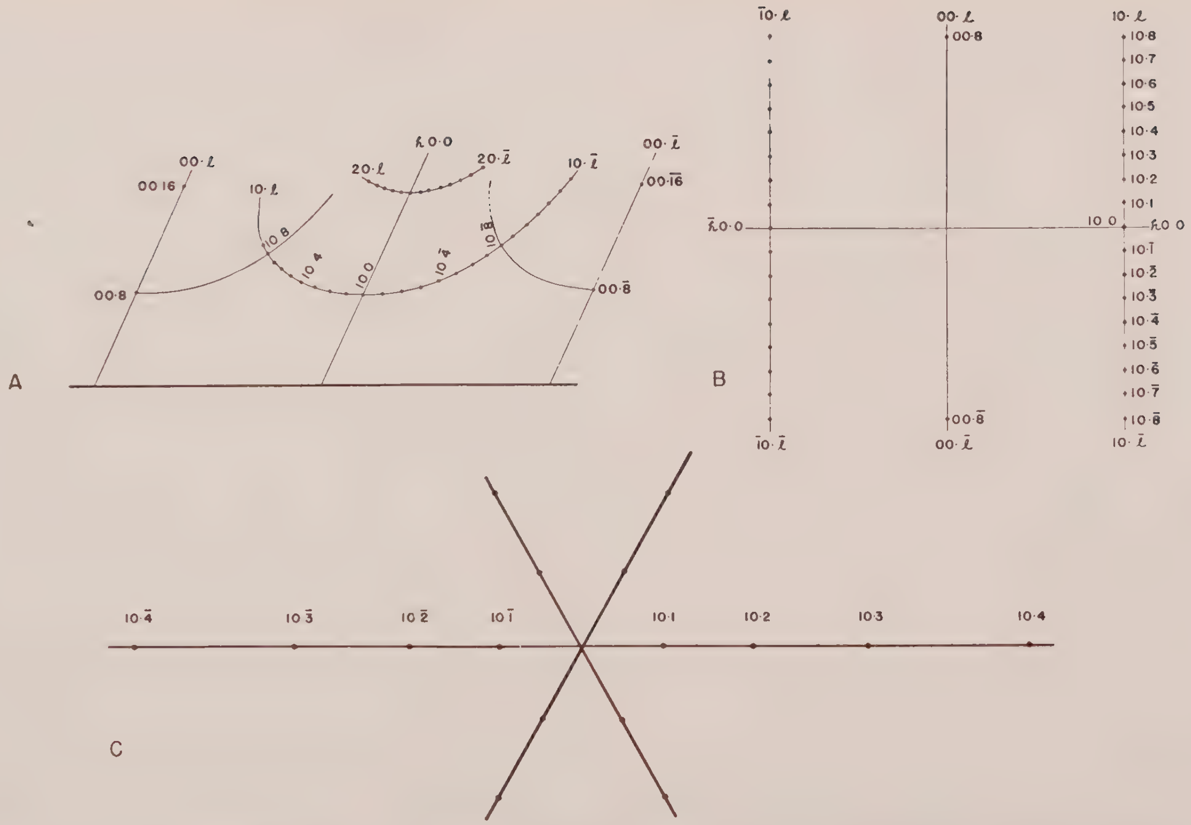

FIG. 4. The relative resolution of $10\cdot l$ reflections for silicon carbide type *8H* by the Weissenberg method (*A*), Buerger precession method (*B*) and Laue method (*C*). Crystal-to-film distance, 6 cm. for Laue and precession films, Weissenberg camera diameter, 5.73 cm.; CuK_α radiation. The over-all figure has been reduced in size.

This method has three disadvantages. Two of these, which have been mentioned above, are not serious. These are the presence of "unwanted" spots, and the limited number of $10 \cdot l$ spots that appear on the regular size flat film. The third disadvantage has to do with the intensities of the spots. One is usually not concerned with this, however, unless he has found a new type and desires to calculate its structure. In this case he can investigate the crystal further using an a axis Weissenberg film or a Buerger precession film showing a^* and c^*.

The writer has not investigated any other polytypic compound but he feels that the principles considered in this paper could easily be applied to other substances of this same nature. It might be mentioned that Frondel and Palache (1950) have published Laue patterns of the zinc sulfide types $4H$, $6H$ and $15R$ and have stated that the forms can be separated conveniently by direct comparison of these films.

The writer wishes to thank Professor L. S. Ramsdell, of the University of Michigan, for his many helpful suggestions, and Mr. Fred D. Pitman, of Wyandotte, Michigan, for his assistance in the preparation of the figures.

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ZONING IN PEGMATITE DISTRICTS*

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Abstract

In numerous pegmatite districts the size, shape, external and internal structure, texture and mineral content of an individual pegmatite can be shown to be functions of the distance of the body from its batholith source. Not all of these characteristics are independent variables, for the host rock is an important factor in influencing shape and external structure and thus indirectly internal structure. It is suggested that mineralogically and structurally complex pegmatites owe their generally distant position from the batholith to a relatively late withdrawal from the pegmatitic hearth with resulting increased fluidity, greater rare-element content and greater penetrability of the surrounding rocks.

Introduction

Widespread and intensive field investigations of pegmatite mineral deposits during the last 15 years have yielded considerable information on their mineralogy, internal structure and genesis. More recently additional attention has been directed toward the study of pegmatite districts or provinces and toward relating the characteristics of individual pegmatites to their regional distribution and geology. Such studies, although yet few in number, already reveal some systematic variations within pegmatite provinces. This *regional zoning* of pegmatites apparently is comparable to zoning that has been recognized in various districts containing hydrothermal mineral deposits, typified, for example, by Butte, Montana (Sales and Meyer, 1949), Cornwall, England (Davidson, 1930), Leadville, Colorado (Laughlin and Behre, 1934) and the Cordillera Real, Bolivia (Turneaure and Welker, 1947). For a general discussion of zoning in hydrothermal districts, reference should be made to the work of W. H. Emmons (1937, 1940), one of the chief advocates of the zoning concept.

The purpose of this paper is to report some new examples of zoning in pegmatite districts, to examine critically others recorded in scattered references and to discuss the factors that influence such zoning.

In preparing this summary the writer has been greatly assisted by discussions with many geologists, particularly L. R. Page and F. S. Turneaure. Special thanks are due to K. K. Landes for a critical reading of the manuscript and for several suggestions toward its improvement. The problem of zoning in pegmatite districts was explored in a preliminary way by J. J. Hayes (1948) at the writer's suggestion. The study also has been advanced materially by a grant from the Faculty Research Fund of the Horace H. Rackham School of Graduate Studies, Univer-

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sity of Michigan, which enabled the writer to study pegmatites in European countries during the summer of 1950.

Development of the Zoning Concept

Recognition of the close spatial ties between pegmatite swarms and marginal parts of batholiths was employed as one of the original arguments for the magmatic origin of pegmatites. DeBeaumont (1847), for example, related the distribution of pegmatites to other characteristic peripheral phenomena ("granite aura") of granite masses. Subsequent writers on pegmatites have underlined the spatial and genetic relationships between offspring pegmatites and parent batholith. Landes (1933, p. 38) summarized viewpoints on this association thus: "Of greatest frequency is the occurrence of pegmatite in the laterally adjacent country rock. However, many pegmatite bodies occur entirely within what is probably the parent intrusive. In this case the contact between pegmatite and enclosing rock may be a gradational one." Emmons (1940, p. 25) also emphasized that, "Many pegmatites are found in the upper parts of granite batholiths and in their roof."

A few early observers presented more detailed information on systematic pegmatite variations within a district. Brögger, who generally was far in advance of his contemporaries in his understanding of pegmatites, described (1890) an excellent example of mineralogical zoning in the Langesundfjord district, Norway. Van Hise (1904, p. 724), pictured the arrangement of Black Hills pegmatites as follows: "From the central mass (of granite) great quartz-feldspar dikes radiate. In passing away from the core the dikes become smaller and have a less typical form; at the same time the material assumes the appearance which we ordinarily denominate pegmatitic. . . . Still farther away the pegmatitic masses begin to have vein-like characters—that is, there is a rough concentration of the material in different layers parallel to the walls. Still farther away a true banded-vein structure is found."

Except for casual and scattered observations, however, the phenomenon of district zoning was not studied further in a systematic way until Gevers (1936) supplied a classic description of the Namaqualand deposits, which he found were zonally arranged in three groups: interior, marginal and exterior. This classification correlated mineralogical and structural variations of the pegmatites with their positions relative to the source batholith. A similar type of zoning to which Gever's divisions were applicable was discovered by the writer (1948) in the Eight Mile Park district of Colorado. Emmons (1940, pp. 23–24) related the zoning to the vertical dimension: "Nearly all pegmatitites that carry metals, lithium minerals, and gems are found in the roof regions of batholiths above the lower limit, or dead line, . . . The quartz-feldspar-mica pegmatites, however, are found also deep within batholiths. They form at higher temperatures than do most of the metalliferous veins and are found lower in the parent masses and farther below the roof contacts than are mineral veins. The dead line for such pegmatites is much deeper that that for metalliferous veins, and in many large bodies of granite and of granite gneiss there seems to be no lower limit for quartz-feldspar-mica pegmatites."

The problem of district zoning has been discussed by Cameron *et al.* (1949, pp. 5–9), who cite a number of examples and conclude that "These examples are enough to indicate that such (district) variations offer a promising field for further and more systematic study."

Discussion of the interrelations of the factors involved is difficult, for we lack quantitative data on pegmatite composition and on changes in the vertical dimension. Nevertheless this review and summation will have achieved its purpose, if it succeeds in provoking its readers into further consideration of the problems advanced.

EXAMPLES

Africa

Namaqualand (Gevers, 1936; Gevers, Partridge and Joubert, 1936). The Namaqualand pegmatites fall into three groups: those in the central part of the granite batholith, the *interior* pegmatites; those within the batholith along its periphery, the *marginal* pegmatites; and those outside the batholith in the metamorphic rocks, the *exterior* pegmatites. Interior pegmatites are abundant, narrow and erratically distributed. They contain chiefly quartz, microcline and some muscovite, garnet, schorl and beryl. Pegmatites of the relatively narrow marginal zone are not as abundant but commonly are larger than the interior type. In their general mineralogy they resemble those of the interior, but some have been extensively altered by post-magmatic mineralization. The exterior pegmatites, which occur in a 10-mile wide zone beyond the batholith borders, characteristically display a well developed internal structure with cores of massive quartz. Many of them have been extensively modified by hydrothermal replacements, involving the formation of albite, muscovite, tourmalines, spodumene, lepidolite, columbite-tantalite, euxenite, polycrase, monazite, xenotime, lithiophilite, molybdenite, bismuth, scheelite and chalcopyrite. It is in these pegmatites that the bulk of the rare-element minerals is concentrated. Farthest from the batholith is the zone of hydrothermal quartz-tourmaline veins.

Erongo, Area, South-West Africa (Gevers and Frommurze, 1929; Gevers, 1942). Interior pegmatites are not reported. Marginal pegmatites that occur along joint planes in the granite and at its contacts usually strike parallel with the metamorphic foliation. They contain microcline, quartz, graphic granite, albite, schorl and rarely some cassiterite. A well developed internal structure occurs in many. The exterior-zone pegmatites, which are in schist, also show internal structural units and generally are tabular or lens-like sills. Schorl is characteristically rare, but many bodies are albitized and contain associated cassiterite, muscovite, lepidolite, alkali tourmaline, triplite, lazulite, molybdenite, arsenopyrite and bismuth minerals.

Kabba Province, Central Nigeria (Jacobson and Webb, 1946, 1947). Within the Old Granite occur interior-type pegmatites containing chiefly microcline and quartz. Those in the marginal zone are similar with some biotite as well. Some are albitized, but they contain only small amounts of columbite-tantalite or cassiterite. In the schists and gneisses, exterior pegmatites of microcline, quartz and muscovite are albitized with cassiterite and columbite-tantalite.

The zonal distribution of wolframite, cassiterite and columbite in Nigeria has been described by Haag (1943), who finds three zones:

(1) Outside the Younger Granite wolframite predominates; cassiterite is minor or absent; columbite is absent.

(2) Within the Younger Granite near its margins cassiterite predominates; wolframite may be abundant; columbite begins to appear.

(3) Deeper within the granite columbite predominates; cassiterite is abundant; wolframite decreases.

Asia

Kodarma, India (Biswas, 1929; Roy, Sharma and Chattopodhyay, 1939). Holland (1902) noted that Indian mica-bearing pegmatites occur mainly in mica schist, whereas pegmatites in granite do not contain mica. In Kodarma two groups of pegmatites have been recognized. Those in the parent granitic gneiss (Dome gneiss) and in closely adjacent schists and paragneisses are microcline-quartz-graphic granite pegmatites with subordinate plagioclase and muscovite, very minor tourmaline, garnet, beryl and apatite and no mineable concentrations of muscovite. These can be classed as marginal-zone pegmatites. Exterior deposits, which are confined to the schists and paragneisses, are typical plagioclase-muscovite pegmatites with microcline rare or absent. Conspicuous accessories are apatite, garnet, beryl and schorl or biotite. The individual exterior pegmatites are markedly differentiated with wall zones of schorl, muscovite, quartz, garnet and apatite, an intermediate feldspathic zone, a core-margin unit rich in muscovite and beryl and a quartz core.

Turkestan Range, U.S.S.R. (Shsherbakov, 1936; Strelkin, 1938). On the northern slope of the Turkestan Range a porphyritic granite of Variscan age has been intruded into middle Paleozoic gneisses, mica schists and andalusite schists. Pegmatites have formed in an aureole around the granite and are especially developed along the south or hanging-wall side of the pluton. Three well-defined zones are recognized: (1) Biotite-schorl pegmatites within the granite close to both the north and south contacts and in the metamorphic rocks as far as 100 meters from the margins. Microcline, quartz and oligoclase are the chief minerals. (2) Schorl-muscovite pegmatites with beryl that occurr in the schists and gneisses in a zone 300–800 meters from the granite. (3) Albitized pegmatites, 800–1500 meters from the granite, only along its hanging-wall side. These contain cleavelandite, spodumene, lepidolite, alkali tourmalines, beryl, amblygonite, lithium-manganese phosphates, columbite and economic concentrations of cassiterite.

Europe

Iddefjord, Norway. Vogt (1931), noted that within the granite "great" pegmatites are rare but small and very small ones are common. Within the surrounding gneisses, "great" pegmatites with internal zones, are common.

Halden (*Fredrichshald*), *Norway* (Brögger, 1906). The pegmatites of the eastern zone, closest to the granite, contain chiefly quartz, microcline and micas, whereas those in the northern part of the district are richer in rare minerals.

Langesundfjord, Norway (Brögger, 1890). The feldspathoidal syenite pegmatites of this district lie mainly within the parent augite syenite and nepheline syenite bodies. The rare mineral content of the pegmatites increases markedly toward the nepheline syenite contacts: The zone 5–3 kilometers from the contact is distinguished by pegmatites bearing anorthoclase, albite, nepheline, sodalite, barkevikite, lepidomelane, magnetite and traces of zircon, apatite and pyrochlore. Those bodies from 3 0 kilometers from the contact contain most of the rare mineral concentrations including moreover, such rare species as eudidymite, hiortdahlite, cappelenite, gibbsite, leucophane, nordenskioldine, meliphane, melanocerite, rosenbuschite, wöhlerite, johnstrupite, hambergite and eudialite. Moreover, most rare species are represented strongly only in pegmatites within 1.5 kilometers from the nearest contact. Brögger's map (1890) outlines the zone of meliphane-bearing pegmatites and is thus the first map showing mineralogical zoning in a pegmatite district.

South America

Bolivia (Ahlfeld, 1936). Pegmatites of the Bolivian tin belt, which contain biotite, muscovite, molybdenite, cassiterite and some wolframite occur within the "roof-zone" of the batholith. Hypothermal types of veins are further out, "within the inner zone of contact" (Ahlfeld, 1936, p. 61).

Eastern Minas Gerais, Brazil. Regional variations have been recorded by Pecora *et al.* (1950, p. 248) as follows: "In general, simple pegmatites characterize the mica districts south of the Rio Doce, whereas complex pegmatites are characteristic of the region north of the river. Certain accessory minerals occur more commonly in specific districts or in geographic groups of pegmatites; for example, phosphate minerals are most abundant in the area north of Conselheiro Pena, tantalite and columbite in the district near Governador Valadares and Poaia, and beryl in the districts near Conselheiro Pena and Governador Valadares." According to Pecora (priv. comm.) the areal geology is too imperfectly known to permit a correlation between these variations and position of the parent batholith, at the present time.

Canada

Yellowknife-Beaulieu Area, Northwest Territory (Jolliffe, 1944; Rowe, 1952). Jolliffe (1944, p. 2) states, "Pegmatites carrying rare-element minerals are most common in the "hot" (i.e. contact-metamorphosed) sediments around bodies of younger granite. Some occur within the border phases of such bodies. . . . So far as is known they do not occur within the "cool" (i.e. unmetamorphosed) sediments." The district zoning has been delineated by Rowe (1952, pp. 10–11), who states, "This (regional) zonation . . . features five zones. The first zone, the zone closest to the granite, contains large, irregularly shaped pegmatites that have poorly developed internal structure, and that contain graphic granite. The second zone is composed of pegmatites containing graphic granite and beryl. In this zone the pegmatites tend to be lenticular and have a somewhat better developed internal structure. The third zone contains many pegmatites that have beryl as a component, but have no graphic granite. These pegmatites are regular in shape, and much smaller, and display a readily recognizable internal structure. The pegmatites of the fourth zone are structurally similar to those of the third but many contain columbite-tantalite as well as beryl. The fifth zone, the one farthest from the granite, features pegmatites that are spodumene-bearing." These zones are outlined in Rowe's figure 1B.

Southeastern Manitoba (DeLury, 1929; DeLury and Ellsworth, 1931). Pegmatites occur both in the granite and in metamorphic roof pendants. The former type contains few rare minerals and no economic mineral concentrations. In the roof-pendant pegmatites, however, rare-element minerals are much more common, including spodumene, lepidolite, amblygonite, topaz, beryl, apatite, monazite, lithium phosphates, columbite-tantalite, uraninite, bismuth, bismuthinite, molybdenite, arsenopyrite and sphalerite. These dikes also are markedly zoned. At West Hawk Lake (Stockwell, 1933) microcline pegmatites are common in volcanic and sedimentary rocks near the granodiorite contact, albite pegmatites predominate farther away and lithium pegmatites lie at a still greater distance from the contact.

Preissac-LaCorne Area, Quebec (Norman, 1945; Tremblay, 1947; Derry, 1950). Pegmatites in the central part of the granite bodies are of the common quartz-microcline-muscovite type with a little garnet. They are in part irregular in shape with weak control by the granite on their emplacement. Those with rarer minerals occur near the margins of the intrusives and commonly show orderly internal structure. The sequence of mineralogical zones outward from the batholith is: (1) Beryl pegmatites with columbite-tantalite. (2) Spodumene-muscovite pegmatites. (3) Molybdenite pegmatites with bismuthinite and pyrite. The zones were mapped by Tremblay (1947, figure 7).

United States

Maine. Bastin (1910) observed that the pegmatites with unusual minerals were concentrated in the border zones of granite batholiths and recognized two mineralogical types: a sodium-lithium group and a fluorine group. In the Poland quadrangle Hanley (1939) found that pegmatites with lithium minerals are farther from the granite contact than the potash feldspar-beryl pegmatites.

West-central New Hampshire (Chapman, 1941; Olson, 1942). Pegmatites within the Mount Clough pluton of the Bethlehem gneiss are segregation, shear zone and filled fissure types. They are normally small and consist dominantly of quartz, microcline, plagioclase and subordinate muscovite, biotite, schorl, garnet and beryl. Pegmatites in schist (Littleton formation) outside the pluton contain, in addition to the above constituents, cleavelandite, triphylite, graftonite, apatite, uraninite, autunite and columbite as well as other accessory species.

Middleton District, Connecticut (Foye, 1922; Cameron and Shainin, 1948). In this district Foye (1922) noted that pegmatites with rarer minerals are confined almost exclusively to the Bolton schist or to the Middleton series around the Monson (Glastonbury) granite gneiss, and contain such species as cleavelandite, beryl, muscovite, lepidolite, cookeite, spodumene, garnet, alkali tourmaline, chrysoberyl, triplite, triphylite, lithiophilite, monazite, zircon, columbite, samarskite, microlite, uraninite, autunite, molybdenite and sphalerite. Subsequent studies by Cameron and Shainin (1948) on the beryl deposits of the district confirm the earlier observations. Their map (p. 355) shows that 10 of the 12 main beryl-bearing pegmatites are either in the Bolton schist or in the Monson gneiss very close to its contacts with the schist.

Spruce Pine District, North Carolina (Maurice, 1940; Olson, 1944). The pegmatites are genetically related to bodies of alaskite. The pegmatites are larger and more abundant near the margins of the alaskite masses than in their interiors, and most pegmatites occur in migmatite, gneiss and schist around the alaskite. In the interior pegmatites biotite is almost absent. Very thick pegmatites are most common near the alaskite border, either in the alaskite or in the adjacent country rock. In these marginal-zone bodies the ratio of microcline to plagioclase probably is higher than in pegmatites farther out. These also contain more green muscovite, much of it with A-structure. Rum and ruby muscovites appear mainly in exterior pegmatites, with the rum type generally closer to the intrusive masses and the ruby type farther out to the west and northwest. The more calcic plagioclase is reported from exterior pegmatites, and the most sodic plagioclase apparently occurs chiefly in marginal-zone bodies.

Franklin-Sylva District, North Carolina (Olson and others, 1946; Heinrich, in press, *A*). Pegmatites within the Whiteside tonalite—granodiorite batholith are small and simple in their mineralogy, containing mainly quartz, oligoclase and muscovite. A zone of pegmatites in gneisses closely adjacent to the batholith on its northwest side and in roof pendants of gneiss is characterized by the presence of deep green to dark brown muscovite, commonly heavily stained by magnetite, and by the absence of much microcline. Accessory minerals, which are rare, include beryl, samarskite, alkali tourmaline, gahnite, zircon, autunite, torbernite, uranophane and fine-grained almandite. Farther out to the northwest the muscovite is mainly red-brown and ruby in color, and the accessory assemblage consists of allanite, pyrite, pyrrhotite, chalcopyrite, bornite, schorl, kyanite and large single crystals of spessartite. Some pegmatites of this zone also contain units in which microcline is a major constituent, and others carry abundant coarse biotite.

Jahns and Lancaster (1950) also have recorded for the Hartwell district of Georgia and South Carolina and the Ridgeway-Sandy Ridge district of Virginia regional color variations in muscovite, in which pegmatites in and near the intrusives contain green mica, whereas in those farther away the mica is rum colored.

Alabama (Hunter, 1944, Heinrich, in press, *B*). Cassiterite-bearing pegmatites occur in feldspathized schist near the margins of the Pinckneyville tonalite. The tonalite plutons are elongated northeast-southwest, and many of the tin deposits occur along their northwestern flanks.

At greater distances from the contacts muscovite pegmatites are strongly developed, containing schorl, garnet, biotite and rare apatite, beryl, tantalite, kyanite, pyrite and graphite. As in the Erongo district of South-West Africa schorl and cassiterite are generally mutually exclusive.

Pikes Peak-Florissant District, Colorado. The Pikes Peak granite in parts of Douglas, El Paso, Teller and Park counties contains groups of small pegmatites in which an outer layer of graphic granite frames a central vug partly filled with microcline and amazonite crystals, clear and smoky quartz crystals, topaz, phenacite, fluorite, zircon, biotite, muscovite, cleavelandite, siderite, cassiterite and additional rarer species. In outer parts of the batholith and in the metamorphic rocks around its periphery the pegmatites contain a markedly different accessory suite: muscovite, biotite, apatite, triplite, beryl, columbite, garnet, magnetite and pyrite and are of larger size and entirely different internal structure.

Eight Mile Park, Colorado (Heinrich, 1948). In the central parts of the Pikes Peak granite body occur swarms of tabular, well-zoned pegmatite dikes that average 1–2 feet in thickness and consist of microcline, quartz, oligoclase, muscovite, biotite and schorl. These interior pegmatites have not been hydrothermally altered. Within the granite along its borders are relatively large, horizontal or gently dipping sheet or discoidal pegmatites that crosscut the primary granitic foliation. They possess a rude internal structure with small core pods and are made up chiefly of microcline, quartz, muscovite, biotite and graphic granite. In a few of these marginal pegmatites post-magmatic mineralization has produced small replacement units of albite, muscovite, beryl, tourmaline, garnet and triplite along the footwall core contacts. The exterior-zone deposits, which are large sills in the Idaho Springs schist, are well-zoned and in many cases have been extensively altered by hydrothermal replacement, with the formation of albite (including cleavelandite), muscovite, garnet, schorl, apatite, beryl, triplite, columbite, lepidolite, alkali tourmaline, natromontebrasite and chalcocite. Nearly all of the commercial feldspar, mica and beryl has been mined from exterior pegmatites.

Micanite District, Colorado (Hanley, Heinrich and Page, 1950; Bever, 1952). Pegmatites of economic significance or of complex mineralogy in the Micanite district are clustered close to the sides of an elongated granodiorite body, which appears to be a large projection from the roof of an underlying Pikes Peak granite batholith. These marginal pegmatites, which occur mainly in the flanking sillimanite and biotite schists and to a much lesser extent in the granodiorite itself, have been mined for feldspar and mica and contain the assemblage: muscovite, biotite, beryl, garnet, apatite, schorl, Fe-Mn phosphates, magnetite, Bi minerals, euxenite, cordierite and sillimanite. Pegmatites at greater distances from the granodiorite are not zoned and lack the accessory mineral suite.

Quartz Creek District, Colorado (Hanley, Heinrich and Page, 1950). Pegmatites in the marginal parts of the granite body and in the hornblende gneiss close to the granite contacts are characterized by a strong lithium replacement phase in which cleavelandite, lepidolite, zinnwaldite, pink muscovite, alkali tourmaline, alkali beryl, topaz, microlite, columbite, monazite, gahnite, fluorite and rarely spodumene and amblygonite were formed. Pegmatites at greater distances from the contacts contain muscovite, beryl, schorl and minor columbite, monazite and samarskite.

Other Colorado Districts. In the Climax area of northeastern Lake County, Butler and Vanderwilt (1933) have noted the marked concentration of pegmatitic material in country rock schist and in granite close to their contact and the dearth of pegmatites in the interior parts of the granite mass. The bodies contain accessory muscovite, biotite, magnetite and garnet, but pegmatites in the granite lack biotite. Crawford (1913) has reported similar relationships in the Monarch district of Chaffee and Gunnison Counties. District zoning of pegmatites in the northern part of the Front Range has been recognized by Boos (1947).

Petaca District, New Mexico (Jahns, 1946). Pegmatites in the granite of this district are of small size, irregular shape, simple mineralogy and homogeneous structure. Those in the metamorphic rocks marginal to the batholith are structurally of two types: (*a*) Sills and dikes parallel with the foliation in strike but crosscutting in dip, and (*b*) other types of dikes. Most of the mica deposits and concentrations of rarer minerals such as fluorite, columbite, monazite, beryl and samarskite occur in the last type.

Boulder Batholith, Montana (Heinrich, 1949). Within the batholith pegmatites are associated with aplitic dikes in which they may occur as pods or tabular masses. These segregation-type pegmatites are commonly zoned inward as follows: aplite, fine-grained graphic granite, medium-grained granitic quartz-microcline pegmatite, vugs with crystals of microcline, quartz (usually smoky), albite and sphene. Other marginal pegmatites have small quartz cores and contain accessory amethyst, allanite, schorl, biotite, garnet, apatite, magnetite and pyrite. The exterior type of pegmatite is normally a sill with variable zonal development. Muscovite and garnet are common in some; others contain rose quartz, schorl, epidote, graphite, apatite, fluorite and zircon.

DISCUSSION

Zone variables

The differences between groups of pegmatites in a single district are due to variations in the following characteristics: size, shape, structural relationship to wall rocks (external structure), internal structure (zoning, etc.), texture and mineral content. Not all of these characteristics are independent variables, nor does their systematic variation in all cases depend exclusively on the relative distances of the pegmatites from their parent batholith.

The discussion is handicapped, of course, by the fact that, in some districts of low relief and where the attitudes of the batholithic contacts are not known, we cannot readily relate pegmatite variation to any factor save *horizontal* distance from an *outcrop* of batholithic rock.

Relation of size and shape to type of host rock

The shape of an intrusive* pegmatite body may be controlled to a very large extent by the structure of the rock into which the pegmatitic magma was intruded. Injections of pegmatitic magma will tend to follow preexisting channelways or potential channelways, namely, fractures, faults, crests, or troughs of folds, foliation planes and contacts between different rock units. In competent or massive rocks, such as granite, granodiorite, quartzite, marble and poorly foliated gneisses, fractures and faults play a major role in localizing pegmatitic intrusion and molding the resulting form of the crystallized body. Thus these pegmatites are commonly discordant and have tabular, anastomosing or entirely irregular outlines. Pegmatites in incompetent rocks, for example the micaceous schists, tend to be at least partly concordant, and are shaped as lenses, tabular sills, inclined tubes, phacoliths, troughs and hoods. These relations have been discussed by Landes (1942), by Cameron *et al.* (1949), and by Staatz and Trites (1950), who also note that, "With the intrusion of large amounts of pegmatitic material, the effect of the country rock on the shape of the pegmatite is usually obscured, and the body assumes an irregular stock like shape. The type of country rock thus appears to have the greatest control over the final shape of the smaller pegmatite bodies."

The average size of pegmatites in a single district seems to be largest in the marginal group. Interior pegmatites usually are small.

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* In the following discussion it is understood that pegmatites are, in the main, of intrusive nature and that wholesale replacement of country rock or segregation and crystallization in situ within batholiths are minor genetic processes.

Relation of size and shape to internal structure

Vlassov (1943) has attempted to define the relationship between (1) internal structure and rare mineral concentration and (2) shape and size. He concludes:

(*a*) That the highest concentrations of rare elements (Li, Cs, Be, Rb, Cb, Ta, Bi, Sn, etc.) are to be found in the largest pegmatite bodies and particularly in those with an oval shape, i.e., lenses and stocks.
(*b*) That zoning is better developed in pegmatites of oval shape than in those of tabular shape.
(*c*) That replacement units are best developed in large oval pegmatites.

These conclusions, although perhaps applicable to certain districts, are untenable for pegmatites of many other regions.

No general relationships between pegmatite size and degree of zonal development have been established. Within a single province some very large pegmatite bodies may be very poorly zoned or unzoned, whereas pegmatites of moderate or even small size display marked internal differentiation. However, in an individual body that pinches and swells, zones normally are more numerous in the thicker parts of the pegmatite, and it is in such bulges that cores or core pods have been developed. As noted by Cameron *et al.* (1949, p. 21), "The core segments generally correspond to these bulges in shape and attitude, and their size commonly is a reflection of the dimensions of the bulge as well." In general in very thin pegmatites, those two feet or less in thickness, zonal structure is rarely developed. This size range of pegmatites has probably the highest percentage of unzoned representatives. But it cannot be asserted, on the other hand, that a large size guarantees a large number of zones.

Similarly it cannot be maintained for pegmatites in general, that those of oval outline (lenses or stocks) show better developed zoning or more strongly developed replacement features than do tabular pegmatites. Among the many outstanding United States examples of tabular pegmatites in which both zoning and replacement units are markedly developed are the Harding pegmatite of Taos County, New Mexico, the Brown Derby pegmatites of Gunnison County, Colorado, and a number of gem and lithium pegmatites of the Pala district, San Diego County, California (Jahns, 1951).

Thus as has been noted in several examples, within a pegmatite district there may be systematic variations in the degree or perfection of internal structural development. These variations may accompany regular differences in shape, but in general, pegmatite shape and degree of development of internal structure appear to be independent features.

Relation of internal structure to mineral content

Unzoned pegmatites normally are mineralogically simple, and those of granitic composition contain microcline, quartz, oligoclase-albite, muscovite, biotite, schorl, spessartite, zircon, apatite and magnetite. Pegmatites that contain zones without secondary petrologic units (replacements or fracture fillings) may have, in addition, such species as low alkali beryl, spodumene, columbite, amblygonite and Fe-Mn phosphates. Pegmatites in which both zones and secondary units occur are mineralogically most complex, and contain cleavelandite, sodic "sugary" albite, lepidolite, zinnwaldite, cookeite, alkali tourmaline, alkali beryl, petalite, pollucite, topaz, tantalite, microlite and sulfides in replacements or fractures fillings. Increasing mineralogical complexity commonly is a function of increasing structural complexity. Large and well-developed replacement units are to be found only in pegmatites in which a magmatic zonal structure was first developed. But of course, the zones may be obscured or partly destroyed by the replacement effects. The formation of zones apparently is prerequisite to the generation of fluids that form the secondary units through reaction and replacement. This relationship weighs heavily in favor of closed system conditions for structurally complex pegmatites.

The questions arise: (1) Are pegmatites with rare element minerals in secondary units of a notably different bulk composition from pegmatites that carry only zones and consist chiefly of the more common minerals? (2) Are unzoned pegmatites of a different bulk composition from zoned deposits of a similar mineral content? The quantitative data available now are inadequate to provide a clear answer. Despite the mineralogical conspicuousness of most replacement units, their total volume is usually small as compared to that of the entire pegmatite body. In an entire district the amount of such hydrothermal material may represent but a minute fraction of all pegmatitic rock. For example, for the Eight Mile Park district of Colorado the volume of secondary pegmatitic material is estimated to be 1% or less of the total pegmatite present. Jahns in describing pegmatites of the Pala district likewise notes (1951, p. 28), "In few of the dikes, for example, do the coarse-grained replacement units appear to constitute more than 1 percent of the total pegmatite material present." Moreover as Gevers (1936, p. 360) has carefully pointed out, replacement does not necessarily require a wholesale transfer of all elements involved but may result from the exchange of only a few ions or radicals. Rare-element replacement minerals are formed if and when these uncommon elements become locally concentrated. In many pegmatites the mechanism for their concentration apparently never began operation, and these constituents are concealed as typical dispersed elements in the more ordinary pegmatite minerals. Examples are Li in muscovite and biotite, Rb and Cs in microcline, muscovite and biotite, rare-earths in apatite, zircon and garnet and Cb and Ta in sphene and zircon. Thus it may be possible that the differences in composition between some complex and simple pegmatites are more mineralogical than chemical and more apparent than real. According to Vlassov (1943) pegmatite shape is an important factor in determining whether the rare elements are accumulated or remain dispersed.

Despite the lack of quantitative compositional data, it is clear that in some pegmatite districts there exist real mineralogical differences among interior, marginal and exterior pegmatites and that these mineralogical variations may also correspond with variations in the internal structure. The causes of differentiation in individual pegmatites are still unknown. Why in a single district some pegmatites develop zones and others of the same general shape and apparent same mineral composition do not, remains to be answered.

Relation of mineral content to type of host rock

The idea that a general relationship exists between wall rock composition and pegmatite composition is an old wives' tale that, despite numerous decapitations, continues to sprout in hydra fashion under succeeding generations of pegmatite investigators. Recent contributors to this notion have been Biswas (1935), Zavaritsky and Kryjanovsky (1937) and Scholz (1950). Exomorphic and endomorphic effects between pegmatites and their wall rocks are exceedingly widespread. However such effects are almost invariably of very limited extent and have not influenced significantly the bulk composition of the pegmatite. It has been suggested that in some cases pegmatite magmas may become rather markedly contaminated through assimilation, e.g., the Quebec type phlogopite deposits (Landes, 1938). If such a process does take place, it is more likely to proceed at depth rather than at the level at which the pegmatite finally crystallizes and where wall rocks may differ from the rocks below. The bulk composition of pegmatites is independent, in general, of the composition of the host rock. In some districts in which pegmatites apparently occur preferentially in one wall rock type, the localization can be ascribed to structural rather than compositional features, e.g., see Jahns (1951, p. 15).

ORIGIN OF DISTRICT ZONING

As stated by Cameron *et al.* (1949, p. 8), "The concept of zonal distribution is appealing in its simplicity, but it may be that factors other than distance are of equal or greater importance in controlling the distribution of types of pegmatite." As noted previously, the pegmatite characteristics that vary within a district and whose systematic variation represent district zoning are size, shape (and external structure), internal structure and mineral composition.

Does the restriction in some districts of small pegmatites to the interior of batholiths mean that at the time of their intrusion only a minor quantity of pegmatite magma was available? Thus interior pegmatites could be interpreted either as coming early in the pegmatite stage when the generation of pegmatite magma had just been begun or late in the stage when the generative process was tapering off. The absence of rare-element minerals in interior pegmatites, however, appears to argue against their late formation and may indicate that this is a pegmatitic fraction that was expelled early, at a time when the rarer constituents had not yet achieved concentration in the batholithic hearth.

The shape of a pegmatite body is largely a function of the structural feature in which it was localized and crystallized, and this feature, in turn, depends considerably on the physical characteristics and tectonic history of the host rock. Wherever the quantity of magma intruded was unusually large, it may have overcome the controlling influence of the guiding structures. Thus shape may or may not be related to distance from the parent batholith.

Internal structure apparently is influenced only to a minor extent by shape and to an even lesser extent by size. The main factor or factors controlling the formation of zones and secondary structural units remain unknown, but in some districts variations in the development of internal units can be systematically correlated with distance from the batholith.

Rare-element mineralogy is controlled to a large extent by the type and quality of internal units. Thus regional variations in structural units are likely to be accentuated by conspicuous mineralogical changes. Yet in some districts there appear to be significant differences in the bulk compositions of the pegmatites of the various zones. In the Spruce Pine and Franklin-Sylva Districts of North Carolina and the Kodarma district of India the amount of microcline varies, so that the pegmatites range in composition from tonalitic through granodioritic to granitic.

Several general hypotheses may be considered in attempting to explain the origin of zoning in pegmatite districts:

(1) It can be postulated that zoning is not directly related to distance from the batholith, but rather that the variable pegmatite compositions are functions of size, shape, and type of wall rock or result from the confusion of two different pegmatite age groups within the same district. The examples cited and the discussion suffice to indicate that this idea is untenable.

(2) Roy, Sharma and Chattopodhyay (1939) believe that the granitic residuum of a batholith splits into K-rich and Na-rich immiscible fractions. The K-rich portion was ejected first while the granite was not completely consolidated. Following complete crystallization of the batholith the increased pressure resulted in expulsion of the Na-rich fraction to a greater distance from the granite. Because the Na-rich pegmatites also contain most of the important mica concentrations, it is suggested that the K for the muscovite is obtained by the Na-rich fluids through attack of the older, potash feldspar pegmatites at lower levels. The happy coincidences required by this hypothesis are sufficient to emasculate it. No microcline pegmatites were found in which the depotassification postulated could be demonstrated. It is reasonable to suppose that, if they existed, at least some would appear near or in the dome gneiss, since that part of the district represented a higher intensity environment, or a deeper or lower "level" in the sense of the authors. Zones cannot be considered in two dimensions alone but must be envisioned as three-dimensional shells. Furthermore the chances are most doubtful that in each case an expulsion of Na-rich fluid would always encounter a previously formed potash feldspar pegmatite, react with it and then be capable of further migration.

(3) Undoubtedly the period of collection and expulsion of pegmatitic rest-magma from a crystallizing batholith is a long one, during which pegmatitic material is released at various intervals depending upon the rate of accumulation and the magmatic pressure. During this lengthy pegmatitic period the batholith is continuing to crystallize and the composition of the residual magma is progressively changed. Thus fluids generated during the latter part of the pegmatitic stage might differ considerably from those formed and released earlier, both in general composition and in the content of rarer elements. This idea also has been advanced by Bateman (1942, p. 53), who stated: "Withdrawals of the early liquid yield simple pegmatite dikes that are varieties of igneous rocks; later withdrawals of a more aqueous stage yield pegmatites, commonly characterized by druses, compounds of tungsten, tin, uranium, titanium, beryllium, phosphorus, chlorine, fluorine, and other elements, . . ." Later Bateman expanded this concept somewhat (1951, pp. 86–87) to say: "The composition of the pegmatites depends in part on the original composition of the magma from which they spring, but also in larger part upon the stage of crystallization at the time they are withdrawn from the magma chamber. . . . If the residual fluid of a granitic magma is tapped off well before the completion of crystallization, it will consist largely of the ingredients of the minerals that are later to crystallize. . . . Such relatively early withdrawals will yield simple pegmatite dikes. . . . If the withdrawal of residual liquid takes place during a more advanced

stage of magma crystallization much of the feldspar will already have crystallized, but particularly there will be a greater concentration of mineralizers, and rare and uncommon constituents."

Vogt (1930, p. 115) thought that the various pegmatites at Langesundsfjord represent various stages of residual magmas "and this may probably be one of the different reasons on which the varying occurrence of the rare (pneumatolytic) minerals, described by Brögger, depends." Raguin (1946) also mentioned this idea of progressive changes in pegmatitic magmas by referring to the simple pegmatites as the "avant-coureurs" of magma and the complex pegmatites as the ultimate residua or quintessence of granitic substance. McLaughlin (1940) also has observed that in the Bridger Mountains of Wyoming complex pegmatites are younger than simple ones.

This hypothesis is an attractive one. If later pegmatitic fractions had higher contents of volatiles as well as rare-elements, their resulting lower viscosities should permit their extrusion to greater distances from the batholith. It might also be maintained that country rocks had by this time been heated to greater distances from the batholith and that the pressure in the magma had been further increased due to complete batholitic crystallization (Morey, 1922). External pegmatites, by this concept, are richer in rarer constituents because their magma was tapped off at the time of maximum maturity of the pegmatitic stage and are further from the batholith because of their lower viscosity, greater pressure and perhaps the extended thermal aureole of the batholith.

Thus the location and composition of a pegmatite depend upon: (*a*) The chemical character of the source batholith; (*b*) The stage of crystallization of the batholith at the time of withdrawal of the pegmatitic material; and (*c*) The penetrability of the rock around the pegmatite hearth.

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STUDIES IN THE MICA GROUP; RELATIONSHIP BETWEEN POLYMORPHISM AND COMPOSITION IN THE MUSCOVITE-LEPIDOLITE SERIES*†

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Abstract

A determination of the crystal structures of about 50 muscovites and lepidolites of known composition has demonstrated the existence of a complete series between muscovite and lepidolite. The polymorphic variation in this series can be correlated with chemical composition, particularly with Li_2O content. A new variant of the muscovite structure has been discovered for which the term *lithian muscovite* is proposed. This modification crystallizes with a structure very similar to that of normal muscovite and apparently links heptaphyllite muscovite with octophyllite lepidolite. Normal musocvite may have as much as 3.3% Li_2O. Micas with 3.4–4.0% Li_2O are generally characterized by poor crystal development and anomalous optical properties owing to their transitional structures. Lepidolites with 4.0–5.1% Li_2O generally crystallize as the 6-layer monoclinic polymorph whereas those with more than 5.1% Li_2O have usually crystallized as the 1-layer monoclinic polymorph. Hexagonal lepidolite is commonly associated with the 1-layer monoclinic polymorph, and both have essentially identical compositions. Twinning may be responsible for the hexagonal form. Polymorphic modifications of lepidolite may occur across sheets and also along the *c*-axis directions in single books. Owing to considerable overlapping of indices and 2V, the various polymorphs may not be distinguished on the basis of optical constants alone.

Introduction

On the basis of chemical evidence it has been suggested by several investigators that a series exists between muscovite and lepidolite. This series is identified particularly by an increase in the Li_2O content, although other chemical changes, such as an increase in SiO_2 and F, and a decrease in Al_2O_3 are also characteristic. However, no serious effort has previously been made to correlate successive chemical changes with the four polymorphs of these micas described by Hendricks and Jefferson (1939). This paper proposes to correlate the successive change in Li_2O content with polymorphism in the muscovite-lepidolite series.

In order to attempt this correlation about 50 specimens of analyzed muscovites and lepidolites with more than 1.5% Li_2O were obtained from about a dozen investigators, museums and laboratories throughout the world. Most of the micas used in this study have been analyzed within about the last 25 years and the results of most have been recorded in the literature. The structures of these, in addition to the structure of about

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450 specimens of unanalyzed muscovites and lepidolites, were investigated primarily by the Weissenberg method. In those cases where crystal development or optical extinctions were poor, the powder *x*-ray method was used. Copper radiation was used throughout.

The first *x*-ray studies on the micas were made by Mauguin (1927, 1928*A*) who measured the units of structure of several varieties of known composition and density. He observed that the *c*-axis of biotite was apparently only half as long as that of muscovite; thus he was the first to establish polymorphism in the micas by means of *x*-ray analysis. Jackson and West (1930, 1933) made the first detailed study of the structure of mica (muscovite) and, in addition, confirmed in greater detail the structure of mica and other layered silicates proposed by Pauling (1930) on the basis of his coordination theory. The most detailed study of the mica group as a whole was conducted by Hendricks and Jefferson (1939) who found numerous polymorphs. In all, seven different polymorphic modifications, embracing 3 crystal systems were found among 100 specimens. In discussing their observations on the lepidolites analyzed by Stevens (1938), Hendricks and Jefferson (1939) stated that there was no evident correlation of composition with the 3 different polymorphs found in these micas. Muscovite they noted was unique among the micas in that it crystallized with but one structure, the 2-layer monoclinic muscovite type. However, Axelrod and Grimaldi (1949) have described a hydrothermal muscovite that has a small, variable 2V and has crystallized with 3 layers in a monoclinic (?) unit cell. This determination is being restudied, but in any case, such muscovite is exceedingly rare and for the purposes of this discussion only the 2-layer muscovite structure (normal muscovite) need be considered.

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Structure of Muscovite

The distinguishing feature of the muscovite structure (space group $C2/c$) is that it is distorted from the ideal mica structure. This is revealed by the presence of certain reflections, $(06l)$ with l odd, which should normally be absent in the ideal arrangement on the basis of the structure factor calculations of Jackson and West (1930, 1933). This distortion results from an incomplete filling of the octahedral positions and is considered by Hendricks and Jefferson (1939) to be the factor permitting only the two-layered structure for muscovite (Fig. 1). Herein also lies the reason why Winchell's grouping of the micas into heptaphyllite and octophyllite divisions is correct, for these terms simply mean that the unit cell contains seven and eight atoms respectively excluding (O, H, and F). Muscovite, $KAl_2(AlSi_3)\,O_{10}(OH,F)_2$, is the type heptaphyllite and phlogopite, $KMg_3\,(AlSi_3)\,O_{10}(OH,F)_2$, the type octophyllite. This relation may also be expressed by saying that the heptaphyllites have only 2/3 of the octahedral positions filled, whereas the octaphyllites have all such positions occupied. The presence of $(06l)$ reflections with l odd must imply departure from the ideal muscovite structure given by Jackson and West (1930, 1933). Hendricks and Jefferson (1939) report these reflections absent in the two-layered biotite-like micas. Thus muscovite (2-layered) has a structure different from that of the 2-layered octophyllite micas.

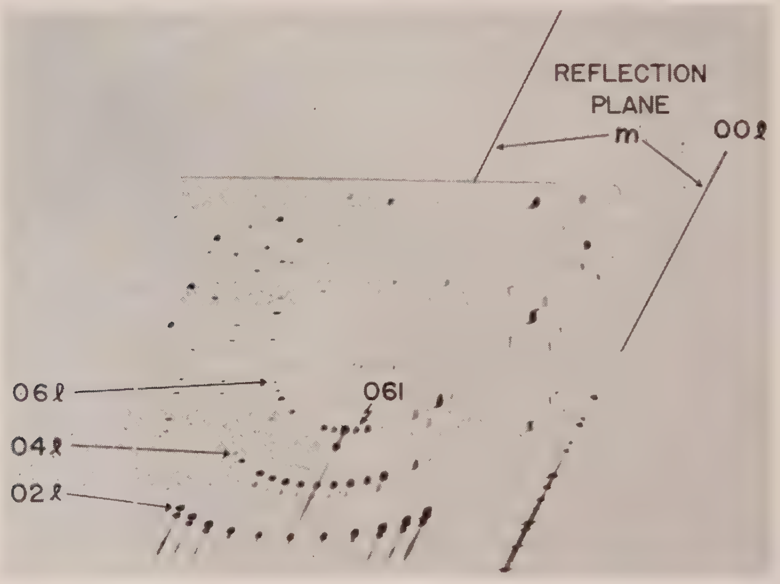

Fig. 1. 0-level a-axis Weissenberg photograph of normal muscovite.

LITHIAN MUSCOVITE

In the course of x-ray studies of so-called lepidolites a new variation of the muscovite polymorph has been discovered. A 0-level a-axis Weissenberg photograph of this form is shown in Fig. 2. It is apparently confined to "lepidolites" with a low Li_2O content and to muscovites with a relatively high Li_2O content. For this variation, the term lithian muscovite is proposed, to distinguish it from normal muscovite. Lithian muscovite is not to be confused with lithium muscovite, the hypothetical end-member used by Stevens (1938) and Berggren (1941). This variety is not common, having been found in only 10 micas of the approximately 500 studied. The following characteristics of lithian muscovite illustrate its close structural similarity to normal muscovite. Both have:

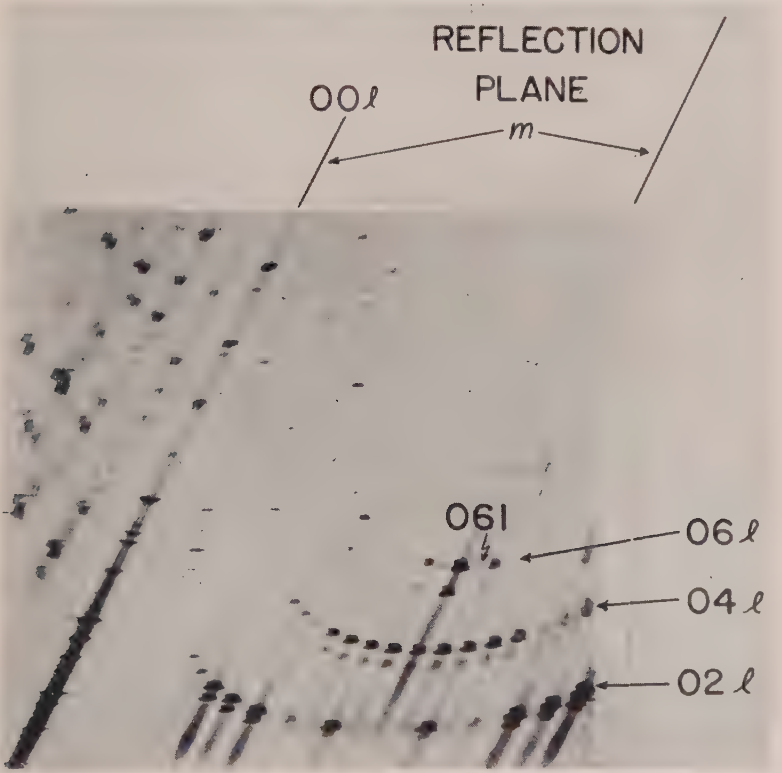

FIG. 2. 0-level a-axis Weissenberg photograph of lithian muscovite.

1. Space group $C2/c$.
2. Cell dimensions (approximate; measured on Weissenberg photographs) $a_0=5.2$ Å; $b_0=9.0$ Å; $c_0=20.0$ Å; $\beta=95°30'$.

3. (06 *l*) reflections with *l* odd present.
4. Optic plane perpendicular to (010).

The following points are different:

1. Indices are in the normal lepidolite range
   $\alpha=1.532$; $\beta=1.552$; $\gamma=1.556$.
2. Several differences occur in intensity of reflections.

The more important intensity differences for (0*kl*) reflections in normal muscovite and lithian muscovite are given in Table 1. The observed intensities of normal muscovite are those of Hendricks and Jefferson (1939). The lithian muscovite structure has been found in pegmatitic micas from: Tördal, Norway; Newry, South Portland and Topsham, Maine; Eight Mile Park, Colorado; Gunnison County, Colorado; Pala, California; Usakos, South West Africa; Kimito, Finland; and Londonderry, Western Australia.

TABLE 1. APPROXIMATE OBSERVED INTENSITIES OF SOME (0*kl*) REFLECTIONS OF NORMAL MUSCOVITE AND LITHIAN MUSCOVITE

Plane	Normal Muscovite	Lithian Muscovite
020	w	a
022	mw	vw
026	a	vw
045	a	vw
061	w	vw
065	vw	vvw
066	w	vvw
067	vw	vvw
069	w	vvw

The data in Table 1 demonstrate that on the basis of the presence of (06*l*) reflections with *l* odd, lithian muscovite must be considered as having crystallized with the muscovite type structure, but with lesser distortion, than in normal muscovite, for most of the (06*l*) reflections with *l* odd recorded are extremely weak. This indicates that lithian muscovite approaches more closely the octophyllite micas in structure and composition for, as Hendricks and Jefferson (1939, page 738) note: these reflections:

> " . . . are absent for the two layer biotite-like micas and none is observed for any of the micas that give (*h*0*l*) intensities of the single layer structure (except muscovite)."

A "lepidolite" No. 1 analyzed by Stevens (1938), which contains only 2.70% Li_2O, has crystallized with the 2-layer muscovite structure, according to *x*-ray analysis by Hendricks and Jefferson (1939). Inasmuch as it contains such a relatively high Li_2O content, Winchell (1942, p. 116) referred to this specimen as a lithium-bearing muscovite. It seemed possible, therefore, that the lithian muscovite structure shown in Fig. 2 was identical with the "lepidolite" analyzed by Stevens (1938) and that Hendricks and Jefferson (1939) neglected to report the intensity differences. Weissenberg photographs of a sample of Stevens No. 1, confirm that this "lepidolite" has the normal muscovite structure as stated by Hendricks and Jefferson (1939). Therefore, Fig. 2, of lithian-muscovite illustrates a new variation of muscovite which must contain at least more than 2.70% Li_2O.

Rowledge (1945) has briefly described a dozen micas and made partial analyses, including Li_2O. *X*-ray studies of portions of these analyzed muscovites and "lepidolites" show that all have crystallized as the normal 2-layer muscovite polymorph. Five of the "lepidolites" have Li_2O contents between 2.17% and 2.60%. Similarly three partially analyzed "lepidolites" from Western Australia described by Murray and Chapman (1931) were *x*-rayed and found to have crystallized with the normal muscovite structure. These micas contain 3.18%, 3.24% and 3.32% Li_2O. This information, coupled with the results obtained from Stevens No. 1 (2.70% Li_2O), and the micas described by Rowledge (1945), substantiates the conclusion that much more lithium may enter the muscovite structure without sensible distortion than has generally been realized.

In a specimen of mica from Newry, Maine, labeled "lepidolite," the lithian muscovite structure grades into that of normal muscovite. The dozen Weissenberg photographs of flakes from this specimen show the critical reflections to vary in intensity between those of normal and those of lithian muscovite. Spectrographic analysis of this material indicated 4.1% Li_2O. A spectrographic analysis of a lithian muscovite from Tördal, Norway shows 4.3% Li_2O. (These quantitative spectrographic analyses are of a preliminary nature. Future wet chemical analyses may indicate they are in need of revision.) Therefore lithian muscovite probably must contain at least 3.3% Li_2O and can contain possibly as much as 4.3% Li_2O.

In attempting to explain a form of muscovite with such a high lithium content, one must inquire if (1) it is possible for such a large amount of lithium to replace aluminum isomorphously in the muscovite structure, and if (2) the increased number of lithium atoms are sufficient to cause a reflection, such as (020), which is present in the muscovite structure, to disappear in the lithian muscovite structure. Since the atomic radii of these elements are similar, $Al = 1.43$Å, $Li = 1.51$Å, the substitution is possible and the valence difference may be countered by other substitutions, such as Fe^{+3} for Fe^{+2}. However, it is not necessary to have isomorphous replacement of aluminum by lithium. Since the muscovite (heptaphyllite) structure has only $\frac{2}{3}$ of its octahedral positions filled, with increasing Li content, the vacant positions gradually can be occupied by additional lithium atoms. This implies the existence of a structural series between heptaphyllite muscovite with a distorted structure, and octophyllite lepidolite with an undistorted structure. The presence of extremely weak (06*l*) reflections with *l* odd in lithian muscovite supports this concept. If these reflections are entirely absent, the mica has the undistorted 2-layer octophyllite type structure.

It is concluded, therefore, that Li ions, in terms of as much as 3.3% Li_2O, can enter the muscovite structure without causing any determinable structural variation. More lithium, probably occupying normally vacant octahedral positions, shifts the structure toward that of the octophyllite micas. In lithian muscovite these changes are already probably of sufficient magnitude to cause a reflection such as (020) to disappear.

Polymorphic Variation in Lepidolite

Most of the specimens analyzed by Stevens (1938), which were structurally investigated by Hendricks and Jefferson (1939, p. 761), were restudied. Table 2 illustrates the high degree of correlation between Hendricks and Jefferson (1939) and the writer's data. The two sets of results are in general agreement. However, some very significant variations may be observed. In Stevens (1938) No. 3 two very small pieces of lepidolite were found from which Weissenberg patterns could be obtained. Both of these gave identical 6-layer monoclinic diffraction patterns. Inasmuch as an optical orientation method is mainly used, the mass of the sample could not be studied by the Weissenberg method because most of the crystals had wavy extinction and gave distorted interference figures. For Stevens No. 6, Hendricks and Jefferson (1939) report the 6-layer monoclinic structure. Two Weissenberg photographs of this sample indicated respectively the presence both of the 6-layer lepidolite polymorph and the 2-layer lithian muscovite type. Therefore, the chemical analysis presented by Stevens (1938) is a composite analysis of the two forms.

A systematic study was made of one book of lepidolite for the purpose of determining the extent of polymorphic variation within a single crystal. Specimen No. 514 from the Opportunity pegmatite, in Gunnison County, Colorado was chosen. The specimen is approximately one and one-half inches across and three-quarters inch thick. It has a uniform typical pink color and is embedded in a mass of cleavelandite with grains of microlite. The sheets are a composite of many crystals. Between many of these crystal units are fine grained aggregates of lepidolite with irregular form and anomalous extinction. The optic planes of the various crystal units in a single sheet are always at 30°, or some multiple thereof, to each other. This appears to be the case regardless of the shape of the crystals in question, and whether or not they are separated by the fine grained aggregate. 2V on all sections of the sheet, regardless of polymorph, remains almost invariant. In all, 27 Weissenberg photographs of small crystals from two sheets, one near the top and the other in the center of the book were made. Of the 14 photographs taken from the sheet near the top, 12 show the 6-layer monoclinic structure and two the 1-layer monoclinic structure. However, on seven of the photographs of the 6-layer monoclinic form, the pattern of lithian muscovite was superimposed, in some cases only faintly present. Of the 13 photographs obtained from crystals in the central sheet, 12 had the 6-layer monoclinic structure and one had the 1-layer monoclinic pattern; only one lithian muscovite pattern was recorded and this again was superimposed faintly on a 6-layer monoclinic pattern. This shows the wide polymorphic variation possible in lepidolite both within sheets and also parallel with the *c*-axis direction. Yet one polymorph, in this case the 6-layer type, is dominant throughout the book. An inspection of Table 2 indicates that except for

TABLE 2. STRUCTURE OF MICAS ANALYZED BY STEVENS (1938)

Stevens (1938) No.	Li_2O	Hendricks and Jefferson (1939)	Levinson
No. 1	2.70	Muscovite	Normal muscovite
No. 2	3.51	Too fine grained for study	Too fine grained
No. 3	3.70		6-layer monoclinic
No. 4	3.81		Too fine grained
No. 5	3.96		Too fine grained
No. 6	5.04	6-layer monoclinic	6-layer monoclinic and lithian muscovite
No. 7	5.05	6-layer monoclinic	not available
No. 8	5.11	single layer	single layer
No. 9	5.33	single layer	single layer
No. 10	5.39	single layer	not available
No. 11	5.51	none available	not available
No. 12	5.64	6-layer monoclinic	not available
No. 13	5.78	single layer	single layer
No. 14	5.89	3-layer hexagonal	3-layer hexagonal
No. 15	6.18	single layer	single layer
No. 16	6.84	single layer	single layer
No. 17	7.26	single layer	single layer

numbers 12 and 14, those lepidolites with more than 5.1% Li_2O have crystallized with the one layer structure. It is necessary therefore to explain the apparently erratic results obtained in the case for No. 12, which has the 6-layer structure and No. 14 with the 3-layer hexagonal structure.

In the case of No. 12, unfortunately no material was available for restudy. It seems possible however, that further investigation could reveal 1-layer forms present in addition to the 6-layer type found by Hendricks. A re-determination of the Li_2O content also would be desirable.

Uniaxial Lepidolite

Uniaxial lepidolite (No. 14) which has crystallized with the 3-layer hexagonal structure has a composition identical with that of lepidolites that have crystallized as the 1-layer polymorph. Such uniaxial lepidolites or nearly uniaxial lepidolites are very rare. A review of the literature revealed that Baumhauer (1903, 1912) first described such material from Mursinsk. He proposed the term microlepidolite for lepidolites of this character in contrast to macrolepidolites with large 2V. No chemical analyses were reported. Uniaxial lepidolite (5.83–6.15% Li_2O) from Western Australia has been described by Winchell (1925), Simpson (1927), Murray and Chapman (1931), Stevens (1938) and Hendricks and Jefferson (1939). This uniaxial lepidolite is associated with a normal biaxial mica of almost identical composition. Winchell (1925, p. 424), in agreement with Simpson (1927), suggests that the uniaxial character "may be due to fine twinning on (001)." Hendricks and Jefferson (1939) reported the structure of uniaxial material from Londonderry as the 3-layer hexagonal type, whereas the biaxial material was the 1-layer polymorph.

Jakob (1927) analyzed uniaxial lepidolite (4.93% Li_2O) from Usakos, and Pehrman (1945) analyzed and described similar material from Kimito (4.99% Li_2O). Specimens of both of these analyzed micas were found to have small areas of a biaxial phase. X-ray studies show that the uniaxial portions have crystallized as the 3-layer hexagonal polymorph but that the biaxial parts have the lithian muscovite structure. Since it has been demonstrated that the lithian muscovite structure is confined to a lower Li_2O range, the Li_2O contents of the Usakos and Kimito micas represent composite results which are lower than those obtainable from strictly uniaxial specimens.

Lundblad (1942, p. 58) notes that an analyzed specimen of lepidolite from Varuträsk has both uniaxial and biaxial portions. The analysis of this material seems unreliable; 65.62% SiO_2 in a lepidolite is too high. Miss Berggren (the analyst), according to Lundblad (1942), seems to suspect quartz contamination. Under these conditions it seems best to eliminate this mica from consideration.

Recently Macgregor (1945) described a lepidolite from Southern Rhodesia with a small 2V. A specimen similar to the described material surrounds a core of muscovite and in turn is enclosed by lepidolite with a large 2V. The muscovite structure is normal, and the lepidolite with the large 2V has crystallized as the 1-layer polymorph, whereas the uniaxial portion has crystallized as the 3-layer hexagonal form. The association of the 1-layer and 3-layer hexagonal polymorphs is identical with that of the Western Australia material. Chemical analyses of these lepidolites are not available.

If one takes into account the presence of lithian muscovite in the Usakos and Kimito lepidolites, it seems reasonable to infer that the Li_2O content of lepidolite of entirely uniaxial character may approach or even exceed the lowest Li_2O content of any of the samples with the 1-layer structure.

Structurally the 3-layer hexagonal polymorph can be geometrically interpreted as three 1-layer forms "twinned" on (001) with succeeding layers rotated 120° in the same direction (either clockwise or counterclockwise). Hendricks and Jefferson (1939, p. 746) with relation to the 3-layer hexagonal structure state that, "a particular layer is repeated about a three-fold screw axis normal to the cleavage." The powder patterns (Fig. 3) of the 3-layer hexagonal polymorph and the 1-layer monoclinic polymorph are almost identical. The cause of this rotation remains to be explained.

Reusch (1869) first obtained an almost perfectly uniaxial figure in muscovite by stacking rotated sheets in this fashion. Although his sheets were of considerable thickness, the over-all optical effect appears to be the same.

Ford (1932) explains the uniaxial effect on the basis of rotation of the plane of polarization. He notes (p. 328):

> "A particularly interesting case, . . . is that of the special properties of superposed cleavage-sections of mica. If three or more of these, say of rectangular form, be superposed and so placed that the lines of the axial planes make equal angles of 60° (45°, etc.) with each other the effect is that polarized light which passed through the center suffers circular polarization, with a rotation to the right or left according to the way in which the sections are built up. The interference-figure resembles that of a section of quartz cut normal to the axis. . . . Further, it is easy from this to understand how it is possible to have in sections of certain crystals (e.g., of clinochlore) portions which are biaxial and others that are uniaxial, the latter being due to an intimate twinning after this method of biaxial portions."

Powder *X*-ray Studies

Hendricks and Jefferson (1939) note that Stevens (1938) numbers 2, 3, 4 and 5, which appear to be in the interval between the 2-layer muscovite type and the 6-layer lepidolite type, are too fine-grained for study and speculate (p. 763):

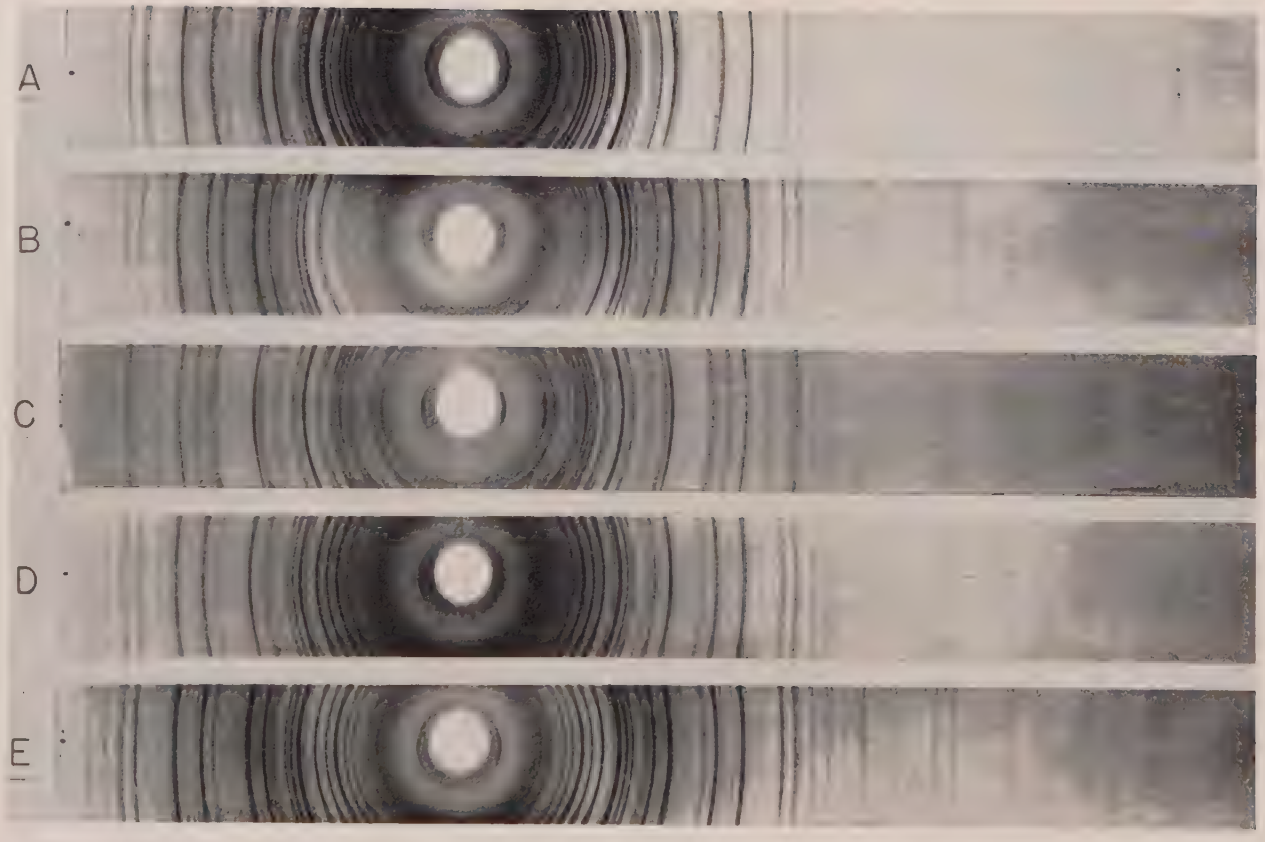

FIG. 3. *X*-ray powder photographs of polymorphs in muscovite lepidolite series. *A*, normal muscovite, Varuträsk, Sweden; *B*, lithian muscovite, South Portland, Maine; *C*, 6 layer lepidolite, Gunnison County, Colorado; *D*, 1 layer lepidolite, Skuleboda, Sweden; *E*, 3-layer hexagonal lepidolite, Londonderry, Western Australia.

> "It is tantalizing to think that samples 2 to 5 owe their poor crystal development to their close approach to the limit of the lepidolite solid solution in muscovite."

It seems very likely that these fine-grained micas owe their macrostructural defects to small-scale variations in their crystal structure which itself probably is assignable to their chemically transitional position in the muscovite-lepidolite series. In order to test this possibility, powder x-ray studies on these micas were attempted.

It was first necessary to obtain powder x-ray data for each of the known polymorphs. The only powder data on muscovites or lepidolites found in the literature were for normal muscovite (Nagelschmidt, 1937) and for the 3-layer muscovite polymorph (Axelrod and Grimaldi, 1949). Grim and Bradley (1951) list partial data for normal muscovite and a 1-layer lepidolite and give photographs of the two patterns. A complete set of powder pictures was compiled by x-raying powdered single crystals whose structure had first been determined by the Weissenberg method. This set of standard photos, which includes the polymorphs of muscovite and lepidolite under study, is shown in Fig. 3; the d-spacings of forms not previously published are given in Table 3.

The structures of the fine-grained lepidolites analyzed by Stevens (1938) have been determined:

Stevens No.	Per Cent Li_2O	Structure by Hendricks and Jefferson	Levinson	
			Weissenberg	Powder
2	3.51	too fine-grained	too fine-grained	6-layer lepidolite+ 2-layer muscovite
3	3.70		6-layer	6-layer lepidolite+ 2-layer muscovite
4	3.81		too fine-grained	6-layer lepidolite+ 2-layer muscovite
5	3.96		too fine-grained	6-layer lepidolite+ (2-layer muscovite?)

That these fine-grained lepidolites are combinations of forms and not single structures confirms the idea that the poor crystal development is related to their composition. Micas with these combined structures may be termed transitional. It is noteworthy that crystals of Stevens No. 3 large enough for Weissenberg photographs are 6-layer forms, but the poorly developed crystals from the same specimen have a combination 6-layer lepidolite and 2-layer muscovite structure. It is likely that the

TABLE 3. SPACINGS OF POLYMORPHIC FORMS
Cu $K\alpha_1$, $\lambda=1.53736$

2-layer lithian muscovite (monoclinic) South Portland, Maine		1-layer lepidolite (monoclinic) Topsham, Maine		3-layer lepidolite (hexagonal) Londonderry, Western Australia		6-layer lepidolite (monoclinic) Gunnison County, Colorado	
I	*d*-spacing	*I*	*d*-spacing	*I*	*d*-spacing	*I*	*d*-spacing
m	9.95	s	9.91	m	9.91	ms	9.89
m	5.01	s	4.98	m	4.97	m	4.99
m	4.50	w	4.94	m	4.66	m	4.49
vvw	3.95	vw	4.34	m	3.84	w	3.84
vvw	3.87	vw	4.12	m	3.58	m	3.61
w	3.71	vw	3.86	vs·	3.31	m	3.47
w	3.46	s	3.61	s	3.09	m	3.31
ms	3.20	vs·	3.33	s	2.86	m	3.19
m	3.21	s	3.07	w	2.651	m	3.07
m	2.98	s	2.87	s	2.571	m	2.88
s	2.84	m	2.675	mw	2.455	m	2.775
w	2.755	s	2.573	mw	2.375	vs·	2.572
ms	2.571	m	2.468	vw	2.243	m	2.416
m	2.474	m	2.387	vw	2.186	vvw	2.248
m	2.387	vw	2.253	mw	2.126	vvw	2.190
vw	2.247	m	2.132	vw	2.052	vw	2.039
vvw	2.196	ms	1.988	m	1.984	s	1.985
mw	2.132	vvw	1.956	vvw	1.955	vvw	1.684
m	2.081	w	1.748	w	1.718	vvw	1.633
vw	1.951	vw	1.715	mw	1.643	vvw	1.572
w	1.742	m	1.646	w	1.611	m	1.506
vw	1.720	vvw	1.581	w	1.576	vvw	1.393
m	1.644	vvw	1.544	w	1.547	vvw	1.355
vvw	1.596	m	1.511	mw	1.511	vvw	1.319
vvw	1.557	vw	1.493	vvw	1.481	w	1.300
vvw	1.510	vvw	1.420	vvw	1.457	vvw	1.239
w	1.500	vvw	1.375	vvw	1.435		
w	1.487	vvw	1.352	vvw	1.411		
vvw	1.453	vw	1.337	w	1.341		
vw	1.427	vw	1.299	vw	1.295		
w	1.340	vvw	1.242	vvw	1.285		
w	1.296	vvw	1.199				
vvw	1.269	vvw	1.136				
vw	1.243						
vvw	1.220						
vvw	1.199						

well developed crystals have a higher lithium content than the intergrown portions. In all cases the 6-layer lepidolite polymorph predominates over the muscovite type in the transitional types.

Owing to the close similarity between the powder patterns of normal muscovite and lithian muscovite the exact type of muscovite in the combined powder patterns of the transitional micas is difficult to determine with certainty. However, careful study of the intensities indicates the probable presence of the lithian muscovite type.

CORRELATION

On the basis of the structural data obtained it seemed advantageous to plot Li_2O content against the polymorphic types of the muscovite-lepidolite series: normal muscovite, lithian muscovite, transitional micas, 6-layer lepidolite, 1-layer lepidolite and 3-layer hexagonal lepidolite. The result of plotting the Li_2O contents of 35 analyzed micas against their structures is presented in Fig. 4. Many more micas with less than 2% Li_2O were studied, but there is no need to record these, as they all have the normal muscovite structure.

The graph shows that micas with less than ca. 3.3% Li_2O have the normal muscovite structure; those with 3.4%–4.0% L_2iO have transitional structures; those with 4.0%–5.1% generally have crystallized with the 6-layer lepidolite structure; whereas those biaxial micas with more than 5.1% Li_2O generally have the 1-layer lepidolite structure. The uniaxial lepidolites as discussed previously have relatively large Li_2O contents, probably in the same range with those of the 1-layer form, and this structure may be explained by "twinning." Too few data are now available to place the lithian muscovite type of structure accurately in the series. However, the available evidence shows that it belongs somewhere between normal muscovite and 6-layer lepidolite. The above conclusions are graphically represented in Fig. 5.

Analyzed micas described by Berggren (1940, 1941) and Lundblad (1942) do not fit well into the conclusions represented by Fig. 5. The results obtained from *x*-ray studies of these micas are presented in Table 4. Inconsistencies appear with regard to Berggren's (1940) micas in *A*, *B*, *C*, and Lundblad's (1942) No. 10 and No. 13. Micas *C* and No. 13, which are reported to have 3.9 and 5.7% Li_2O, respectively, have crystallized with the normal muscovite structure. As much as 5.7% Li_2O, or even 3.9% Li_2O, in normal muscovite appears improbable on the basis of present knowledge. It is possible that the lithium content of the mica samples varies so much that the portions supplied represent extreme structural variations of the analyzed materials. Several of the Swedish micas are optically non-homogeneous, which may indicate the presence of several crystal phases and one may even be contaminated with quartz (Lundblad, 1942). Under such circumstances an attempt to correlate polymorphism with chemistry would be fruitless. Baumhauer (1912) described several lepidolites with a wide range of 2E and several distinctive types of etch figure in single specimens. Both of these features probably indicate structural variations. Except for the Swedish micas the correlation is good; indeed better than might be expected, considering the presence of structural variation in single books and the probable wide range in quality of the Li_2O determinations.

LEGEND FOR FIGURE 4

1. Wodgina, Western Australia; No. 36, Rowledge (1945).
2. Tabba Tabba, Western Australia; No. 52, Rowledge (1945).
3. Varuträsk, Sweden; Analysis D, Berggren (1940).
4. Wodgina, Western Australia; No. 6, Rowledge (1945).
5. Tabba Tabba, Western Australia; No. 6, Rowledge (1945).
6. Karibib, South Africa; 2.5% Li_2O, courtesy of Foote Mineral Company.
7. Wodgina, Western Australia; No. 2, Rowledge (1945).
8. Manitoba, Canada; No. 1 Stevens (1938).
9. Karibib, South Africa; 3.0% Li_2O, courtesy of Foote Mineral Company.
10. Wodgina, Western Australia; Murray and Chapman (1931).
11. Tabba, Western Australia; Murray and Chapman (1931).
12. Ravensthorpe, Western Australia; Murray and Chapman (1931).
13. Bear Claim, Manitoba; Spec. XXXIII, Ellsworth (1932).
14. Pala, Calif.; No. 2, Stevens (1938).
15. Pala, Calif.; No. 3, Stevens (1938).
16. San Diego Co., Calif.; No. 4, Stevens (1938).
17. Pala, Calif.; No. 5, Stevens (1938).
18. Ubini, Western Australia; Murray and Chapman (1931).
19. Poona, Western Australia; Murray and Chapman (1931).
20. Pala, Calif.; No. 6, Stevens (1938).
21. Ohio City, Colorado; No. 7, Stevens (1938).
22. Pala, Calif.; No. 12, Stevens (1938).
23. San Diego Co., Calif; No. 8 Stevens (1938).
24. Mesa Grande, Calif.; No. 9, Stevens (1938).
25. Antsongombato, Madagascar; Mauguin (1928*B*).
26. Wakefield, Canada; No. 10, Stevens (1938).
27. Mesa Grande, Calif.; No. 13, Stevens (1938).
28. Maharitra, Madagascar; Mauguin (1928*B*).
29. San Diego Co., Calif.; No. 15, Stevens (1938).
30. Antsongombato, Madagascar; No. 16, Stevens (1938).
31. Greenland; No. 17, Stevens (1938).
32. Usakos, Southwest Africa; Jakob (1927).
33. Kimito, Finland; Pehrman (1945).
34. Calgoorie, Western Australia; No. 14, Stevens (1938).
35. Londonderry, Western Australia; Murray and Chapman (1931).

The structures of all the above micas, with the exception of numbers 21, 22 and 26, have been determined by the author. In the case of the three exceptions the structures reported by Hendricks and Jefferson (1939) have been used.

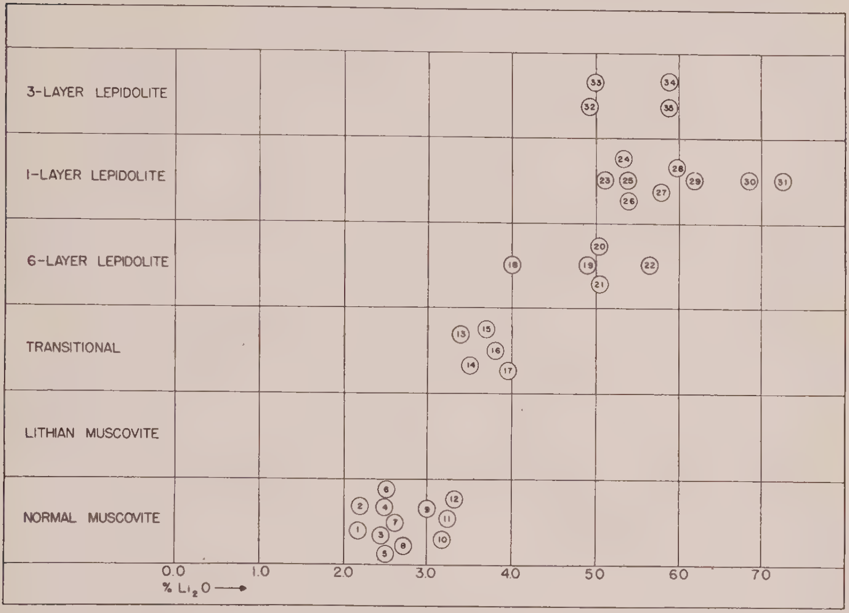

FIG. 4. Plot of the Li_2O contents of 35 micas in muscovite-lepidolite series against the various polymorphs.

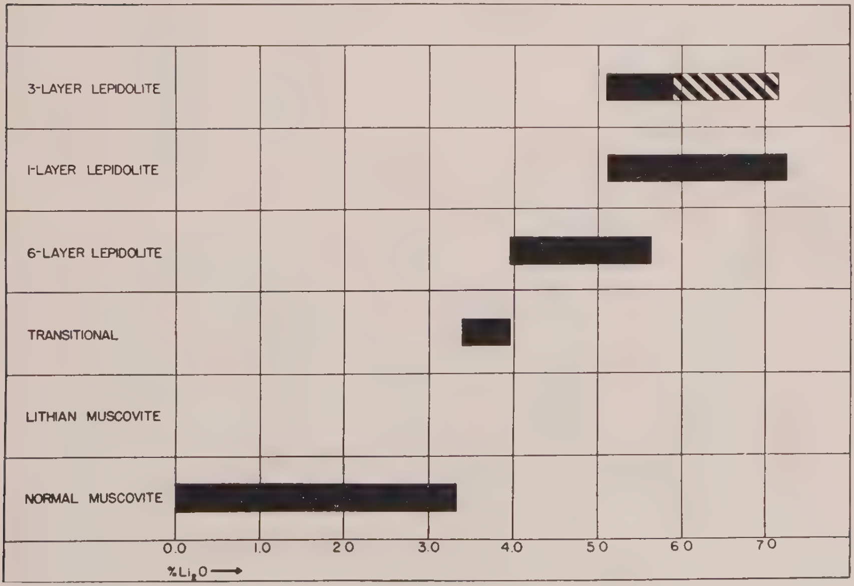

FIG. 5. Idealized representation of relationship between Li_2O content and polymorphism in the muscovite-lepidolite series.

TABLE 4. STRUCTURE OF MICAS DESCRIBED BY BERGGREN (1940, 1941) AND LUNDBLAD (1942)

Analysis	% Li_2O	Structure	Locality
A	5.95	6-layer lepidolite	Varutraäk
B	4.35	1-layer lepidolite	Varuträsk
C	3.9	normal muscovite	Varuträsk
D	2.45	normal muscovite	Varuträsk
E	1.80	normal muscovite	Varuträsk
G	0.73	normal muscovite	Varuträsk
H	0.69	normal muscovite	Varuträsk
I	0.22	normal muscovite	Varuträsk
J	0.76	normal muscovite	Varuträsk
K	1.10	normal muscovite	Varuträsk
L	1.1	normal muscovite	Varuträsk
No. 10	4.55	1-layer lepidolite	Varuträsk
No. 13	5.7	normal muscovite	Utö
No. 14	5.5	6-layer and 1-layer lepidolite	Rozena

OPTICAL DATA

Optical constants were measured on individual flakes whose structures first had been determined by the Weissenberg method. The results are presented in Table 5. The indices of refraction were measured on the Abbé refractometer or were obtained by the immersion method and are reproducible to ±0.001. The Mallard method was used in the determination of 2E and 2V was calculated. The only other determinations of the optical properties of lepidolite polymorphs have been by Hendricks and Jefferson (1939). Their results, though scanty, are in close agreement with those of this paper.

The lepidolite polymorphs are not distinguishable on the basis of optical properties alone; it follows therefore their general chemistry cannot be deduced from the optical constants. A trend is that the indices, and particularly 2V, of the 1-layer polymorph tend to be slightly higher than those of the 6-layer polymorph, but the overlap is broad. This can be accounted for by the optical inactivity of the Li-ion. Although variations in Li content influence the layer stacking pattern, changes in optical constants are more sensitive to variations in Fe^3, Ti, Fe^2 and Mn, both in muscovite and the lepidolites. The indices of lepidolite vary within books (note values obtained from spec. 514 and 452 in Table 5). However the indices of lithian muscovite are in the range of normal lepidolites whose values are considerably below those of normal muscovite.

TABLE 5. INDICES OF REFRACTION AND 2V OF LEPIDOLITES AND LITHIAN MUSCOVITE*

Specimen Number	Locality	α	β	γ	2V
	Six-layer lepidolite				
502	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.555	1.560	30°
514 (2)	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.553	1.557	30°
514 (3)	Opportunity Pegmatite, Ohio City, Colorado	1.533	1.555	1.560	33°
514 (4)	Opportunity Pegmatite, Ohio City, Colorado	1.533	1.557	1.560	33°
514 (5)	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.555	1.558	—
514 (6)	Opportunity Pegmatite, Ohio City, Colorado	1.531	1.553	1.560	33°
505 (*b*)	Brown Derby Pegmatite, Ohio City, Colorado	1.530	1.556	1.560	34°
967 (*a*)	Stewart Mine, Pala, California	1.530	1.552	1.559	33°
967 (*b*)	Stewart Mine, Pala, California	1.529	1.554	1.558	34°
970 (*a*)	Stewart Mine, Pala, California	1.534	1.555	1.559	33°
452 (*a*)	Varuträsk, Sweden	1.532	1.556	1.561	33°
452 (*b*)	Varuträsk, Sweden	1.535	1.556	1.561	35°
452 (*c*)	Varuträsk, Sweden	1.532	1.552	1.557	37°
	Three-layer hexagonal lepidolite				
539 (*f*)	Pope mining claim, Southern Rhodesia	—	1.568	1.568	0°
539 (*g*)	Pope mining claim, Southern Rhodesia	—	1.566	1.566	0°
	One-layer lepidolite				
514 (*k*)	Opportunity Pegmatite, Ohio City, Colorado	—	1.556	1.558	—
464 (*b*)	Newry, Maine	—	1.556	1.558	50°
471 (*b*)	South Portland, Maine	1.533	1.553	1.560	41°
476 (*b*)	Skuleboda, Sweden	—	1.562	1.564	—
476 (*c*)	Skuleboda, Sweden	—	1.564	1.566	37°
535 (*c*)	Dogon Daji, Nigeria	—	1.557	1.562	47°
539 (*a*)	Pope Mining Claim, Southern Rhodesia	—	1.562	1.565	38°
539 (*b*)	Pope Mining Claim, Southern Rhodesia	—	1.562	1.566	37°
	Two-layer lithian muscovite				
556 (*b*)	Tördal, Norway	—	1.553	1.556	34°
556 (*c*)	Tördal, Norway	1.532	1.554	1.558	34°
465 (*a*)	Newry, Maine	1.533	1.555	1.559	34°
465 (*f*)	Newry, Maine	1.534	1.555	1.561	37°
471 (*a*)	South Portland, Maine	1.529	1.551	1.556	32°
679 (*a*)	Usakos, S. W. Africa	1.534	1.559	1.566	—

* Measurements by Mr. Charles H. Hewitt.

Specimens numbered such as 415 (2) through 514 (6), or 452 (*a*) through 452 (*c*) represent measurements obtained from different flakes in the same book.

SUMMARY

The discovery of the lithian muscovite type of structure, the interpretation of the structural origin of the 3-layer hexagonal lepidolite polymorph and good correlation between Li_2O content and the various polymorphs shows that chemical and structural transitions occur between Li-free muscovites and Li-rich lepidolites. It seems probable, however, that the relationship between a single ion variation and polymorphism (Fig. 5) in a structure as complex as the micas is somewhat oversimplified. Overlapping undoubtedly takes place and it may be possible to get even more accurate results by calculating the number of atoms with octahedral coordination against the various polymorphs; at present too few complete analyses and specimens are available to attempt this approach.

The study shows also that structural variations in single lepidolite crystals are common and can be coupled with compositional variations. Optical data are insufficient to distinguish the various polymorphs. The correlation may prove valuable in estimating the approximate Li_2O content of the mica without a chemical analysis.

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DIRECTIONAL GRINDING HARDNESS IN DIAMOND*

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ABSTRACT

In order to measure the relative variation of grinding hardness in single diamond crystals, two mechanisms have been constructed. One, which might be called a hardness comparator, permits the simultaneous cutting of two crystals. One crystal is oriented in an arbitrarily chosen unit hardness direction. The cutting rate of the second crystal is compared with the cutting rate of the reference crystal.

To facilitate the measurements a second apparatus has been designed to prepare spherical surfaces of predetermined radii on the diamonds. This apparatus was used in connection with hardness variation measurements on the cube face. The remainder of the planes investigated were studied on pyramidally faceted stones, because of the very slow operation of the sphere grinder.

Hardness curves were plotted, hardness against grinding azimuth, for the cube, dodecahedron, and octahedron, and for intermediate planes in the zones containing these three planes.

Hexoctahedral symmetry was assumed since no evidence of lower symmetry has been obtained from hardness measurements.

Errors inherent in the method are discussed.

The fact that certain directions in a diamond may be readily cut with diamond dust on a cast iron wheel while other directions can not be cut or polished is well known to the diamond cutting trade. A thorough qualitative investigation has been carried out by Slawson and Kohn.[1] The purpose of this research is to compare quantitatively the abrasion rates on single crystal diamonds for a series of planes in the three principal crystallographic zones.

If a diamond is cut in a relatively easy grinding direction ("on grain" in the professional cutter's parlance), the diamond itself tends to replenish the abrasive and the wheel will remain in good cutting condition almost indefinitely. If, on the other hand, a hard or "off grain" direction is cut, the abrasive is not replenished and the quality of the lap surface deteriorates rapidly. When this occurs, the diamond is no longer cut and the wheel becomes badly scored. In order to minimize the effects of this variation, two diamonds were cut simultaneously. One was used as a reference diamond. Its orientation was unchanged during the entire study. The other diamond was oriented so that special planes in the three principal zones were ground. A series of azimuths across each of these planes was investigated. The two diamonds were mounted on spindles provided with graduated azimuth circles. To avoid scoring the lap and to assure that both diamonds were ground under nearly equal conditions, they were moved back and forth on opposite sides of the center along a diameter of the lap. The thrusts of both diamonds on the lap surface were the same. In most tests the thrust was maintained at two kilograms, although in a few instances lighter thrusts were used. The relative rates of removal of material from the two stones provided a measure of the relative grinding hardness for each direction under consideration. Four micron diamond dust in olive oil was used for all grinding operations. Figure 1 shows the slide bar supporting two spindles over the wheel. The cam which oscillates the bar may be seen below the table in the left foreground.

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* Contribution from the Department of Mineralogy and Petrography, University of Michigan, No. 175.

[1] Slawson, C. B., and Cohn [sic], J. A., Maximum hardness vectors in the diamond: *Industrial Diamond Review*, **10**, June, 1950, 168–172.

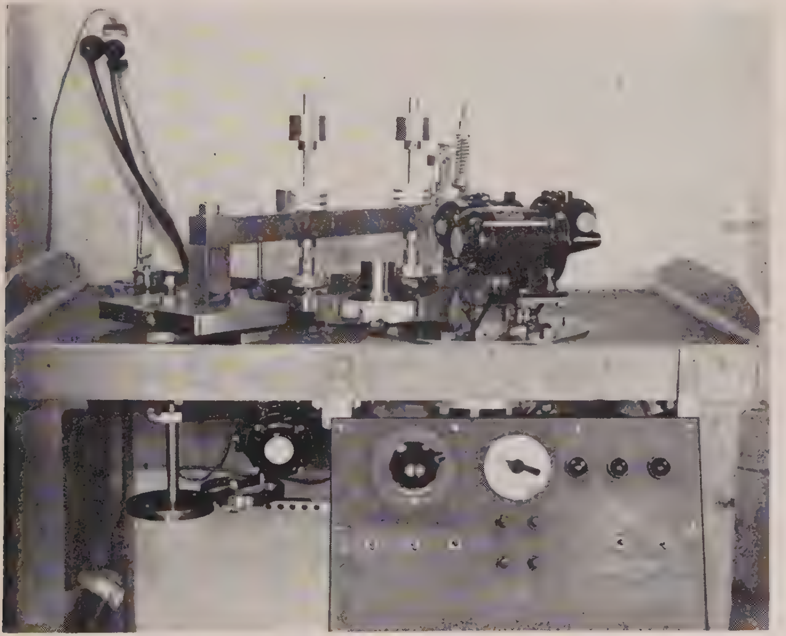

FIG. 1. Diamond hardness comparator and sphere grinder.

A grinding constant, k, was determined for each cut. The relation used was $k = v/TF$, in which v represents the volume of diamond removed; T, the time of grinding; and F, the thrust force exerted upon the lap. A value of k was determined for both the reference diamond and the diamond under test. If k_1 represents the grinding constant of the test diamond and k_2 represents the constant of the reference diamond, then H, the relative grinding hardness, equals k_2/k_1. This hardness is reciprocal to the relative ease of grinding. It should be noted that the values of H are purely relative and that they are not to be considered as indicative of the grinding hardness on an absolute scale.

The hardness of the diamond crystal in some crystallographic directions is equal to, or greater than, the hardness of the diamond dust particles, which presumably are oriented with the octahedral plane parallel to the lap surface. Therefore, when one of the directions being compared is a hard direction of the diamond, small absolute hardness differences may cause large variations in grinding rate.

As a reference direction of assumed unit grinding hardness, the cube face in the direction of an *a*-axis was chosen. This relatively easy cutting direction tends to reduce the damage to the lap when directions are compared, because, as the cube surface is worn away, it replenishes the lost abrasive. Also a sufficiently large advance of cut is assured to minimize the effects of error of measurement. In addition, this direction is readily oriented on the crystals of octahedral habit used in this investigation.

The determination of the volume of diamond removed by grinding necessitates the measurement of the areas of the surfaces before and after grinding and also the measurement of the depths of the cuts. Because of the extreme hardness of diamond, it is not feasible to measure the depth directly. In order to measure the depth and the area of the cuts, it was decided to generate spherical surfaces on the diamonds. Then, the radius of curvature being known, the areas and depths could be readily determined by measuring the diameters of the flats.

An apparatus has been designed to grind the spherical surface. The diamond is mounted on a rotating spindle, oscillated about a horizontal axis, which lies above the lap surface at a distance equal to the radius of the sphere being ground. The spindle is spring-loaded so that it can slide parallel to its length. This sliding motion is limited in the direction toward the lap by an adjustable stop. The diamond should be bounded by a spherical surface when the sliding motion rests against the stop. Were it not for certain extremely hard grinding directions on certain planes, such an arrangement would generate a spherical cap. For each element of the surface of the sphere there are one or more optimum grinding directions, and there may be directions that can not be ground. In order that each point on the sphere be cut in all directions, the whole apparatus was mounted on a third (vertical) axis. A 90° oscillation about this third axis insures that all points on the sphere will be ground in a large number of directions. In order to grind a large number of directions on each element of the spherical surface, these three motions must have an irrational relationship with each other. Furthermore, the third axis oscillation makes it possible to utilize most of the wheel surface. The surface of the cutting wheel tends to be grooved at the innermost and the outermost points where the wheel grinding radius is momentarily unchanged during the end- and mid-points of the third axis oscillation. To lessen this difficulty the outer and inner parts of the wheel are beveled so that the diamond does not rest on the wheel during the time of reversal.

In order to generate a spherical surface, it is necessary first to approximate the surface with a series of small flats. For grinding a 50° cap on the cube position, the grinding process requires about 80 hours, during which time the lap must be resurfaced several times, even though two diamond flats in optimum grinding position are cut simultaneously. Surfaces prepared in this manner showed profiles constant in radius to about 10 per cent. Because of the great amount of time required, only the cube face was studied with the aid of the spherical surfaces. Figure 2 is a photograph of the sphere grinding apparatus.

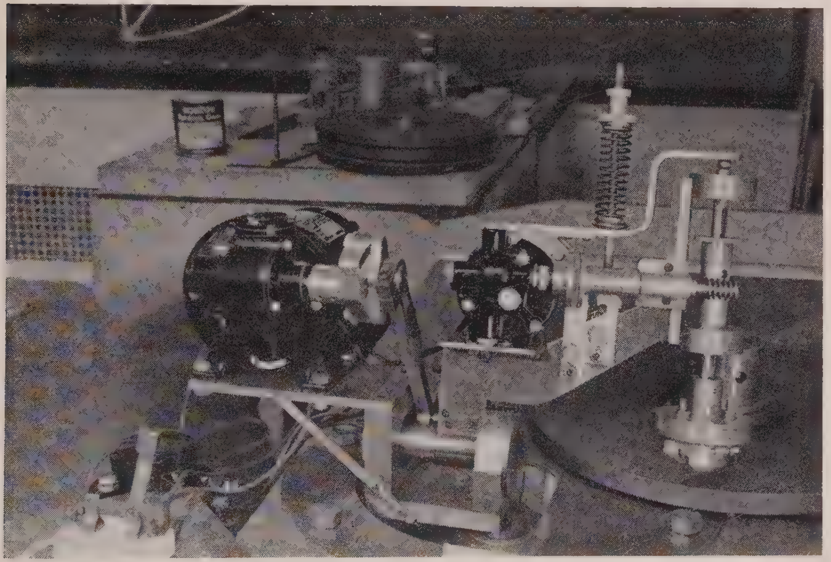

FIG. 2. Diamond sphere grinder.

Planes other than those parallel to the cube were studied from abraded frustrums of pre-formed pyramids. The pyramids were so shaped that the surfaces being abraded had rectangular outlines. Measurements made in this manner are consistent with those made with the aid of spherical surfaces. These ground surfaces of the diamonds were examined under a reflecting microscope and the facets were drawn with a camera lucida. An ocular micrometer, previously calibrated, was used to determine the scale of each drawing. In each instance it was found necessary to focus the microscope critically so that the image of the surface fell exactly in the plane of the micrometer scale. Otherwise parallax introduced serious errors. By this means the slight changes in scale of the drawings due to unavoidable changes in projection distances were measured. The areas were determined with a planimeter. Depths of cuts were determined from the changes in edge lengths of the ground facets.

The diamonds used were about $1\frac{1}{2}$ carat, clear yellow, exceptionally well crystallized, nearly ideal octahedrons with smooth plane surfaces. Because of the fine quality of the surfaces, the crystals could be oriented on the goniometer with a high degree of accuracy. They showed no external evidence of twinning and possessed low double refraction. As the study progressed, it became apparent that some twinning was present, even in these specially selected crystals. The twinned portions of the crystals appeared as occasional fine lamellae on the lapped surfaces parallel to the trace of an octahedral plane. Since the grinding rate on the surface of the twinned areas differs from that of the main untwinned portion of the crystal, a polishing relief is developed. Because of the slight undulations present on some of the polished surfaces, on planes nearly parallel to the octahedron, the twin lamellae appeared as discontinuous crescentic arcs.

The twinning caused uncertainty of the measurement of some of the "softer" grinding directions. Even a very small twinned area which presents a hard grinding direction will seriously retard the advance of a cut on what should be a fast cutting direction. Where possible, twinned areas were avoided.

The variation of the lap condition is not serious in the study of the directions of low grinding hardness. However, in the case of the directions of high grinding hardness, the effect is of considerable importance. As the wheel surface deteriorates, the cutting rate along relatively hard directions is greatly reduced by this condition. The reference diamond which is oriented in an on-grain position will continue to be cut on a relatively poorly charged wheel. Ultimately, if the wheel is not continuously recharged, even the reference diamond fails to advance. The effect can be reduced to a minimum by simultaneously cutting several additional stones in on-grain positions. During the testing of the hard directions, the surface of the lap must be liberally supplied with olive oil and diamond dust. The usual effect of the result of lap deterioration is an apparent increase in relative grinding hardness. The magnitude of this effect is uncertain.

Certain grinding directions are particularly sensitive to orientation errors. An error of 30 minutes in the location of the cube plane will seriously disturb the four-fold symmetry of hardness vectors which that plane exhibits. Undoubtedly the general experience of cutters that only one direction on the cube facet will present optimum cutting conditions[2] is due to small errors in the location of supposedly cube facets. Similar statements could be made concerning other planes. To avoid such errors it is necessary to check the position of facets after each grinding operation to make sure that the diamond has not changed position in its dop during grinding.

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[2] Grodzinski, Paul, Diamond and gemstone industrial production methods: Part XIII, *Industrial Diamond Review*, **9**, 154 (1949).

The relatively soft directions cut rapidly enough so that measurable increases in area can be obtained in a few minutes of grinding. However, in the case of the harder directions, even though the grinding continues for up to two-hour periods, the increment of area is so slight as to be close to the limit of accuracy of the measurements. Therefore, it was impossible to obtain satisfactory measurements for the hardest directions. It is for this reason that the curves are dotted in the region of greatest grinding hardness. The high hardness values have a probable error of about 25 per cent. The low values have a probable error of 10 per cent or less. In computing the probable errors some extreme values were eliminated. These extreme values were associated with prominent twinned areas or poor wheel condition.

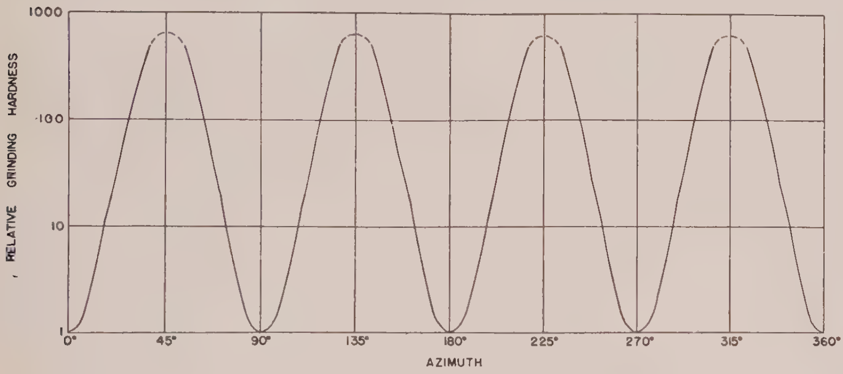

FIG. 3. Cube.

The results of the tests are shown in a series of curves. The curves are based upon measurements in one hundred lapping directions distributed among the several faces. The values were duplicated in successive tests. In the more difficult cutting directions, five or more determinations were made. Orientation of the planes was checked frequently, both by the goniometer and by grinding rates. Grinding rates are accurate checks only when symmetry relationships require that they be equal. The curves are plotted as relative grinding hardness (on a logarithmic scale) against azimuth.

Figure 3 shows the curve for the cube plane. The zero for the azimuth scale is the grinding direction parallel to a crystallographic axis. This direction was chosen as one of unit hardness. Measurements were made at 10-degree intervals up to 40° azimuth. At 45° the cutting is so slow (or the wheel surface deteriorates so rapidly) that no satisfactory measurements can be obtained for this, one of the hardest directions. The 40° azimuth possessed the greatest hardness that could be measured with reasonable certainty.

The extrapolation from 40° to 45° is purely conjectural. All of the directions of the crystal which yielded reasonably consistent results showed the hardness to be a continuous rather than a discontinuous property. Since in general hardness seems to be a continuous property, the extrapolations on this and other faces are drawn so as to avoid discontinuities in the curves. Any alternative extrapolations that involve discontinuities seem unlikely. If there exists a real discontinuity in the curves—assuming that good measurements might be made in the very hard directions in the future—it is a result of the technique employed. Were a harder abrasive available, the apparent discontinuities should disappear.

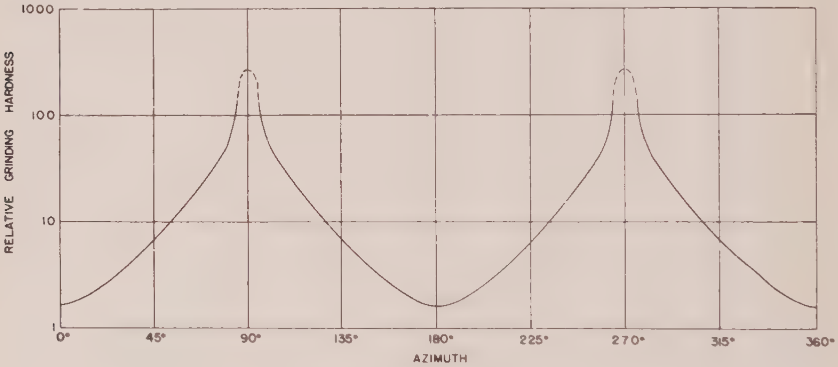

FIG. 4. Dodecahedron.

The only other plane studied that shows hardness magnitudes of the same order is the dodecahedron. The relative grinding hardness curve for the dodecahedron is plotted in Figure 4. The zero azimuth is taken as a direction parallel to a crystallographic axis. In the optimum cutting direction (toward the octahedron), this plane shows a hardness only slightly in excess of the reference direction. When the grinding is toward the cube (parallel to the direction of the long diagonal of a dodecahedron face), the relative grinding hardness is several hundred-fold that of the soft direction. It must be remembered that this does not imply a variation in absolute hardness of such a magnitude. The average rate of change of hardness per degree of azimuth is only about half that of the cube face. No satisfactory measurements were made between azimuths of 80 and

100 degrees. An attempt was made to compare the hardest directions of the cube and dodecahedron directly. Since such tests were inconclusive, it remains in doubt as to whether the cube or dodecahedron possesses the hardest of all known surfaces with respect to grinding.

A curve for the octahedron is not shown as all of the measurements made on this plane, because of its excessive hardness, are unsatisfactory. In general, relative hardness values between 10 and 100 times that of the reference direction were found. These values seem low. The error is tentatively assigned to unfavorable variation in the grinding condition of the wheel. While all of the directions on this face are very hard, none is so hard as the hardest directions on the cube and dodecahedron.

Slawson and Kohn[3] found the maximum hardness to be toward the cube, while the present study showed inconclusively that the direction toward the dodecahedron is the harder. No evidence of the assymmetry reported by Tolkowsky[4] has been encountered either by Slawson and Kohn or by the present writer.

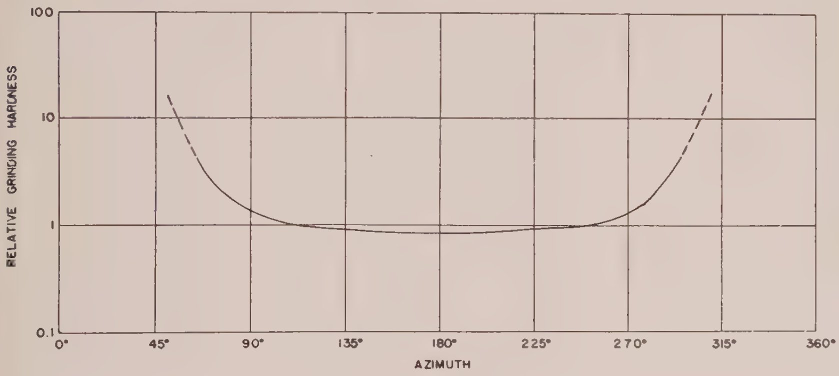

FIG. 5. Zone [010] 23° from Cube.

The octahedron is especially sensitive to orientation errors. If the plane being ground is one degree from the true octahedron position, the hardness curve will show quite low values, in part. In such a case the curve will indicate the orientation error, rather than the approximate hardness variation on the octahedron itself. In the present research the orientation error was kept within 15 minutes. Slawson and Kohn deduced the qualitative relative hardness indicated above by observing the hardness on a series of planes close to the octahedron in the zones

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[3] Slawson and Cohn [sic], *op. cit.*

[4] Tolkowsky, M., Research on the abrading, grinding, or polishing of diamond: (*Unpublished Doctoral Dissertation, University of London,* 1920), cited by Paul Grodzinski, *op. cit.*, Part XII, pp. 118–124.

$[\bar{1}01]$ and $[1\bar{1}0]$. This indirect method probably leads to more reliable results than the direct method of attempting to grind on the octahedron used by the present writer.

The hardness curve for an intermediate plane 23 degrees from the cube in the [010] zone is illustrated in Figure 5. The direction toward the cube is chosen as zero azimuth. This face showed high relative grinding hardness for azimuths up to about 45 degrees. From azimuth 90 to 180 degrees, the face is very easily ground. Such a plane offers the cutter a large range of easy grinding azimuths. The measurements on the hardest directions could not be duplicated well enough to warrant extending the curve to zero azimuth.

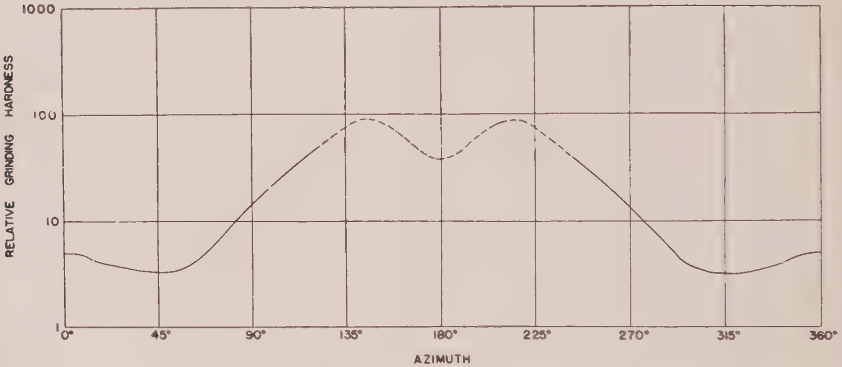

Fig. 6. Zone $[1\bar{1}0]$ 39° from Cube.

In the $[1\bar{1}0]$ zone the results on a plane 39 degrees from the cube are shown in Figure 6. All of the azimuths can be ground, but some only with difficulty. The zero azimuth is toward the cube. The optimum cutting direction lies at azimuth 45 degrees. This direction does not cut so readily as the optimum directions on the previously discussed planes, with the exception of the octahedron. The maximum hardness was found at about 145 degrees. While this represents a very hard direction, the diamond can be cut in this position. The face, obviously, can be cut more readily toward the cube than toward the octahedron. This plane is of interest since it lies at the edge of one of the lapping limits of Slawson and Kohn.[5]

Figure 7 shows the relative grinding hardness curve of a plane 15 degrees from the dodecahedron in the $[\bar{1}01]$ zone. On this plane all directions can be cut. The direction toward the dodecahedron is the softest, while the direction of maximum hardness lies about 100 degrees from the former.

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[5] Slawson and Cohn [sic], *op. cit.*

From the above series of examples it can be seen that it is not sufficient to define the relative grinding hardness merely in terms of the grinding direction within the crystal. For a given grinding direction, the relative grinding hardness depends upon the position of the plane being ground. It is for this reason that the relative grinding hardness can not be expressed as a three dimensional surface analogous to the optical indicatrix.

It should be emphasized that while all of the azimuths on the octahedron face are very hard, the hardest directions are on the dodecahedron plane toward the cube and on the cube toward the octahedron, and that these two hardest directions are parallel to each other. Which of the two latter directions is the harder is uncertain.

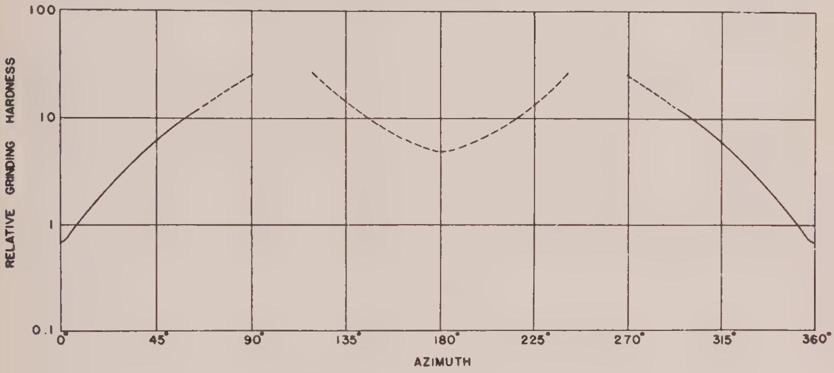

Fig. 7. Zone [$\bar{1}$01] 15° from Dodecahedron.

Acknowledgments

The writer wishes to thank Professor Chester B. Slawson of the Mineralogical Laboratory, University of Michigan, who suggested the problem and general methods of procedure, and, in addition, gave valuable advice during the progress of the study.

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STRUCTURAL CRYSTALLOGRAPHIC RELATION BETWEEN SODIUM SULFATE AND POTASSIUM SULFATE AND SOME OTHER SYNTHETIC SULFATE MINERALS*†

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ABSTRACT

A study of the structural crystallographic relations between the following sulfate compounds has been made using *x*-ray methods as the main tool: Na_2SO_4-K_2SO_4, Na_2SO_4-Li_2SO_4, K_2SO_4-Li_2SO_4, K_2SO_4-$(NH_4)_2SO_4$ and Na_2SO_4-$(NH_4)_2SO_4$.

Aphthitalite, a sodium potassium sulfate mineral, has been prepared in the laboratory from aqueous solutions as well as from fused melts of various sodium and potassium sulfate proportions. *X*-ray methods reveal that at high temperatures close to fusion, sodium sulfate and potassium sulfate are isomorphous and form a complete series of solid solutions. The same synthetic mineral, prepared from aqueous solutions, has been found by *x*-ray investigation and checked by chemical analysis to have a limited range of solid solution varying between the ratios of 1K:1Na to 5K:1Na when crystallized at 70° C. and limited to the almost invariable ratio of 3K:1Na at room temperature.

X-ray powder data are given for $LiKSO_4$ and $LiNaSO_4$, and also for the NH_4KSO_4 member of the continuous series of crystalline solid solution $(NH_4)_2SO_4$-K_2SO_4. It is shown by the *x*-ray powder method that complete immiscibility exists between $(NH_4)_2SO_4$ and Na_2SO_4 at 70° C.

For $LiNaSO_4$ $c_0=9.76$ Å± 0.02, $a_0=7.64$ Å± 0.02, space group $P31c$, $Z=6$, g. (determined) $=2.515$, g. (calc.) $=2.527$; basal twinning with 0001 as twinning plane is common.

INTRODUCTION

The study of the structural relation between sodium sulfate and potassium sulfate is of particular interest in the field of crystallography because of the fact that each compound occurs in more than one crystallographic modification (polymorph) and that there has been some controversy as to whether there exists a solid solution relationship or isomorphism, or both, between certain polymorphs of the two compounds. The existence of such solid solution or isomorphism, if any, is of special interest in structural crystallography because of the big difference in ionic radii between sodium (0.98 Å) and potassium (1.33 Å), which is equal to 0.35 Å or 35.7% of the smaller ion (sodium). Sodium and potassium are not generally grouped together in isomorphous series. Potassium is usually grouped with rubidium and cesium but not with sodium. Tutton (1922) in working out the isomorphous relationship between the alkali sulfates excluded sodium sulfate from his series.

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† Extracted from a thesis submitted in partial fulfillment of the requirements for the degree of Doctor of Philosophy, University of Michigan, June 1952.

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Aphthitalite (or glaserite) has for a long time been assigned the composition $3K_2SO_4 \cdot Na_2SO_4$ or $K_3Na(SO_4)_2$. It has been shown by a number of investigators, Bücking (1889; Douglashall region in Germany), Teeple (1921; Searles Lake, California), Kurnakov, *et al.* (1936; Ural-Emba District, Russia), and Kurnakov, *et al.* (1938; Permian Sea and the salt lakes of Western Kazakhstan, Russia), to be widespread in saline deposits of lake basins. The mineral occurs also around volcanoes, e.g. Vesuvius, Scacchi (1875) and Bellanca (1942). Common among all these investigators, with the exception of Bellanca (1942), is the invariable composition $3K_2SO_4 \cdot Na_2SO_4$ which they assigned to the mineral aphthitalite.

The composition and structure of aphthitalite have been the subject matter of argument and discussion since 1891. The problem is: Is there any crystallographic relation between sodium sulfate and potassium sulfate? Retgers (1891) stated that there is no isomorphism between sodium sulfate and potassium sulfate. He considered aphthitalite as a double salt of the composition $3K_2SO_4+Na_2SO_4$. Van't Hoff (1903), on the other hand, regarded aphthitalite as an isomorphous compound of the not closely investigated hexagonal modifications of both components. Gossner (1904) considered aphthitalite as a binary compound of a constant chemical composition, $NaK_3(SO_4)_2$. Van't Hoff and Barschall (1906) pointed to the existence of a series of solid solutions including the composition $NaK_3(SO_4)_2$. Druzhinin (1938), investigating the system Na_2SO_4-K_2SO_4-H_2O at the temperature of 25° C., considered aphthitalite as a solid solution of the definite chemical compound $K_3Na(SO_4)_2$ with Na_2SO_4 and reported that the compound permits substitution of Na for K within the range $K/Na = 2.44$ to 3.00. Perrier and Bellanca (1940) and Bellanca (1942) using both thermal analysis technique and *x*-ray powder photographs believe that the compound is not $K_3Na(SO_4)_2$ but rather $KNaSO_4$. This forms solid solutions with K_2SO_4 (up to 73 mol per cent total content of the latter) and with Na_2SO_4 (up to 75 mol per cent). Bredig (1942) states that aphthitalite should be considered simply as a solid solution of the high temperature forms of both potassium sulfate and sodium sulfate. Frondel (1950) gives the formula for aphthitalite as $(KNa)_3Na(SO_4)_2$. Winchell (1951) considers aphthitalite, $NaK_3(SO_4)_2$, as "a double salt—not a crystal solution." He mentions that the Na:K ratio seems to vary from about 1:4 to about 1:1.

Sodium sulfate exists in five polymorphous modifications: I, (II), III, (IV), and V. Using the *x*-ray powder method, Kracek and Ksanda (1930) showed that Na_2SO_4 I is stable above 240° C., Na_2SO_4 III is metastable below 185° C. but inert when dry at ordinary temperatures, and Na_2SO_4 V is stable at ordinary temperatures. Polymorphs II and IV are unstable at ordinary pressure. Na_2SO_4 V is equivalent to the naturally occurring mineral thenardite, a rare salt lake deposit and also found near some volcanoes.

Potassium sulfate exists in two polymorphous modifications, namely: (1) a high-temperature modification, called alpha-K_2SO_4, stable above 590° C. and is uniaxial negative, and (2) a low-temperature modification called beta-K_2SO_4, stable at room temperature, orthorhombic, with the space group *Pmcn*. Arcanite is the name of the mineral having the composition K_2SO_4 (equivalent to beta-K_2SO_4) and is said to be of very rare occurrence (Frondel, 1950).

Table 1 is a summary of the crystallographic data of the various sulfates mentioned above.

TABLE 1

Substance	System	Space Group	a_o	b_o	c_o	Z	Ref.
Na_2SO_4							
I	Hex.		5.39		7.25	2	1
III	Orth.	*Pbnn*	5.59	8.93	6.98	4	2
V	Orth.	*Fddd*	5.85	12.29	9.75	8	3
K_2SO_4							
α	Hex.		5.71		7.85	2	4
β	Orth.	*Pmcn*	5.77	10.06	7.52	4	5
$NaK_3(SO_4)_2$ Aphthitalite	Hex.	$P\bar{3}m$	5.65		7.33	1	6
$NaKSO_4$	Hex.	$P\bar{3}m$	5.643		7.259	2	7

1. Calculated from data of Kracek and Ksanda (1930).
2. Frevel (1940).
3. Zachariassen and Ziegler (1932).
4. Bredig (1942).
5. Strukturbericht (1937).
6. Gossner (1928).
7. Bellanca (1942).

THE RELATION BETWEEN SODIUM SULFATE AND POTASSIUM SULFATE

The System Na_2SO_4-K_2SO_4 (*Fused samples*)

Samples were prepared by fusing mixtures of Na_2SO_4 and K_2SO_4 ranging in molecular ratios from 4:1 to 1:4. These gave very similar powder photographs, and it seems apparent that when crystallizing from a melt, there is a continuous solid solution between the hexagonal forms of Na_2SO_4 and K_2SO_4. The pure end members always invert to a lower symmetry modification upon cooling. On the other hand, there is a range of compositions between the end members in which the hexagonal form of the solid solution is stable. On either side of this stable range, as the end member is approached, the originally formed solid solution unmixes, yielding the pure end member, which inverts on cooling, and a stable solid solution phase containing a lower percentage of the end member than was present in the melt.

The limits of this range in which the solid solution is stable are difficult to determine in samples prepared from melts. The material is so fine grained, and the variation in refractive indices so slight, that optical differentiation of a pure end member and a solid solution close to that end member is impractical. The *x*-ray powder photographs of samples approaching the end members in composition show increasing evidence of a phase consisting of the inverted end member. But in the range of 3:1 to 1:3, or even 4:1 to 1:4, this unmixing and inversion is negligible. This difficulty in establishing definite limits for the stable range in material prepared by fusion is not encountered when using single crystals prepared from solution. These are discussed later.

The lattice constants, a_o and c_o, for some members of this high-temperature solid solution series of aphthitalite, have been determined from the corresponding powder photographs. These are given, together with the published data on Na_2SO_4 I (calculated from data of Kracek and Ksanda, 1930) and α-K_2SO_4 (Bredig, 1942), in Table 2. This table shows clearly the increase in the unit cell dimensions as the result of the substitution of potassium for sodium.

TABLE 2

	Na_2SO_4 I	Na_4K	Na_3K	NaK	NaK_3	NaK_4	α-K_2SO_4
a_o	5.39	5.49	5.50	5.64	5.66	5.67	5.710
c_o	7.25	7.26	7.265	7.27	7.33	7.39	7.85

Thin sections and crushed fragments showed aphthitalite to be uniaxial positive. The values of ω and ϵ increase with higher content of potassium. They range from 1.485 to 1.493 for ω, and from 1.492 to 1.500 for ϵ (Na light; values ± 0.002). Winchell (1951) gives the range for ω as 1.487 to 1.491, and for ϵ 1.492 to 1.499, in good agreement with the values found for the synthetic material. The general trend of increase of indices is shown graphically in Fig. 1.

Using the pycnometer method a general increase in the specific gravity was found in going from the high-sodium members to the high-potassium members. The determined specific gravity values range from 2.68 (for

Na_4K) to 2.71 (for NaK_4) with an experimental error of ±0.01. Winchell (1951) gives the specific gravity of 2.7 for aphthitalite of the composition $NaK_3(SO_4)_2$. Gossner (1928) gave the value of 2.697 for the same composition. The specific gravity of aphthitalite from Vesuvius was reported by Bellanca (1943) to be 2.697. The determined specific gravities were found to be in general agreement with those calculated according to the increasing lattice constants given in Table 2.

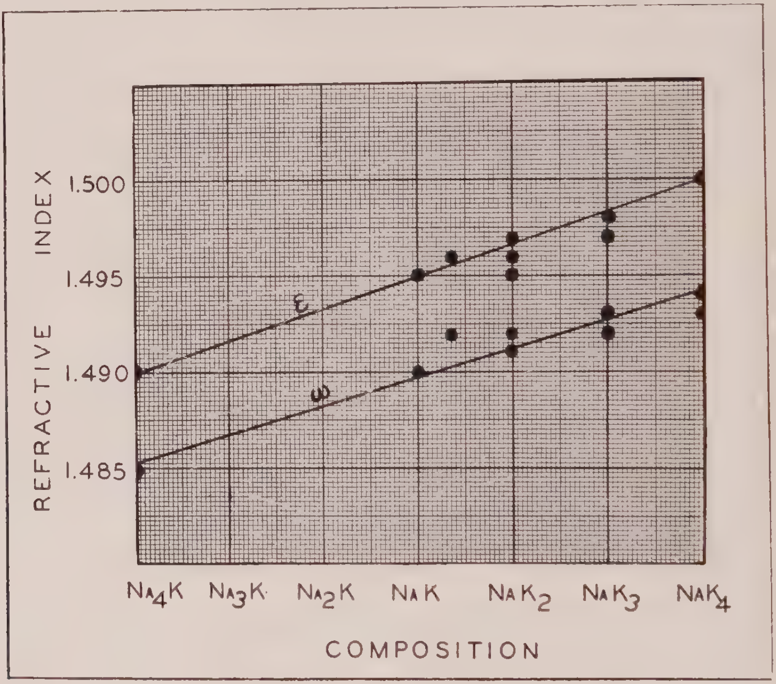

FIG. 1

The System $Na_2SO_4-K_2SO_4-H_2O$ (*Aqueous solutions*)

X-ray powder photographs of crystals grown from aqueous solutions of various molecular ratios of sodium sulfate and potassium sulfate proved to be similar to those taken for the fused aphthitalites. Also powder photographs of crystals obtained from aqueous solution and then fused were found to be similar to those of the unfused ones. This indicates that the crystals grown from aqueous solutions at the temperatures of 70°, 50°, 35° and 20° C. are anhydrous. Weissenberg photographs of these crystals showed the hexagonal symmetry and yielded the space group of $P\bar{3}m$, in agreement with that given by Gossner (1928) and Bellanca (1943). The powder photographs of the various crystals, formed at the same temperature but grown from solutions of different composition show either the aphthitalite type of pattern or that of the end members, Na_2SO_4 V or β-K_2SO_4. It is quite apparent that the crystals do not have the same chemical composition as that of the solution from which they crystallize. Table 3 shows the results of the chemical analyses in terms of the percentage of potassium in each crystal and the corresponding composition of the crystal in terms of the molecular percentage of the two end members. The chemical analyses, made by the periodate method (Willard, Boyle 1941), are within the range of experimental error of ±1.0%. The values in Table 3 were then used in the construction of Fig. 2 which shows the relationship between the composition of the crystals and that of the solutions from which they were grown at the various temperatures of 70°, 50°, 35°, and 20° C. The results of the chemical analyses are in agreement with those of x-rays.

TABLE 3

Composition of Solution		Temperature	K determined quantitatively	Type of x-ray powder pattern	Composition of Crystals Produced	
Na_2SO_4 :	K_2SO_4				Na_2SO_4 :	K_2SO_4
%			%		%	
75.0 :	25.0	70° C.	—	Na_2SO_4 V	100.0 :	—
66.7 :	33.3	70° C.	28.0	Aphthit.	42.0 :	58.0
50.0 :	50.0	70° C.	31.6	Aphthit.	33.0 :	67.0
33.3 :	66.7	70° C.	39.2	Aphthit.	12.9 :	87.1
25.0 :	75.0	70° C.	45.1	β-K_2SO_4	— :	100.0
20.0 :	80.0	70° C.	45.1	β-K_2SO_4	— :	100.0
66.7 :	33.3	50° C.	34.6	Aphthit.	26.0 :	74.0
50.0 :	50.0	50° C.	35.8	Aphthit.	23.0 :	77.0
33.3 :	66.7	50° C.	40.5	Aphthit.	12.0 :	88.0
25.0 :	75.0	50° C.	45.5	β-K_2SO_4	— :	100.0
20.0 :	80.0	50° C.	45.5	β-K_2SO_4	— :	100.0
66.7 :	33.3	35° C.	34.0	Aphthit.	27.0 :	73.0
50.0 :	50.0	35° C.	35.8	Aphthit.	23.0 :	77.0
33.3 :	66.7	35° C.	40.6	Mixture	11.0 :	89.0
25.0 :	75.0	35° C.	45.0	β-K_2SO_4	— :	100.0
20.0 :	80.0	35° C.	45.0	β-K_2SO_4	— :	100.0
66.7 :	33.3	20° C.	35.6	Aphthit.	24.0 :	76.0
50.0 :	50.0	20° C.	36.4	Aphthit.	22.0 :	78.0
33.3 :	66.7	20° C.	45.5	β-K_2SO_4	— :	100.0
25.0 :	75.0	20° C.	45.5	β-K_2SO_4	— :	100.0
20.0 :	80.0	20° C.	45.5	β-K_2SO_4	— :	100.0

The range of the limited solid solution series of aphthitalite, formed from the pure compounds dissolved in distilled water, is shown in Fig.

3, based on the results of both chemical analyses and powder photographs. It is interesting to note that the range of the solid solution decreases with decrease in temperature until it finally shrinks to a very small range at 20° C. At this temperature aphthitalite acts like a compound of fixed composition, and explains why many previous investigators had so regarded it. Perrier and Bellanca (1940) stated that aphthitalites of volcanic origin occur in nature with the ratio of K_2SO_4 to Na_2SO_4 varying from 3:1 to 1:3. When formed from aqueous solutions, on the other hand (artificially or in salt deposits), it always shows the ratio approximately 3:1. This last statement seems to be true only at temperatures of 30° or lower.

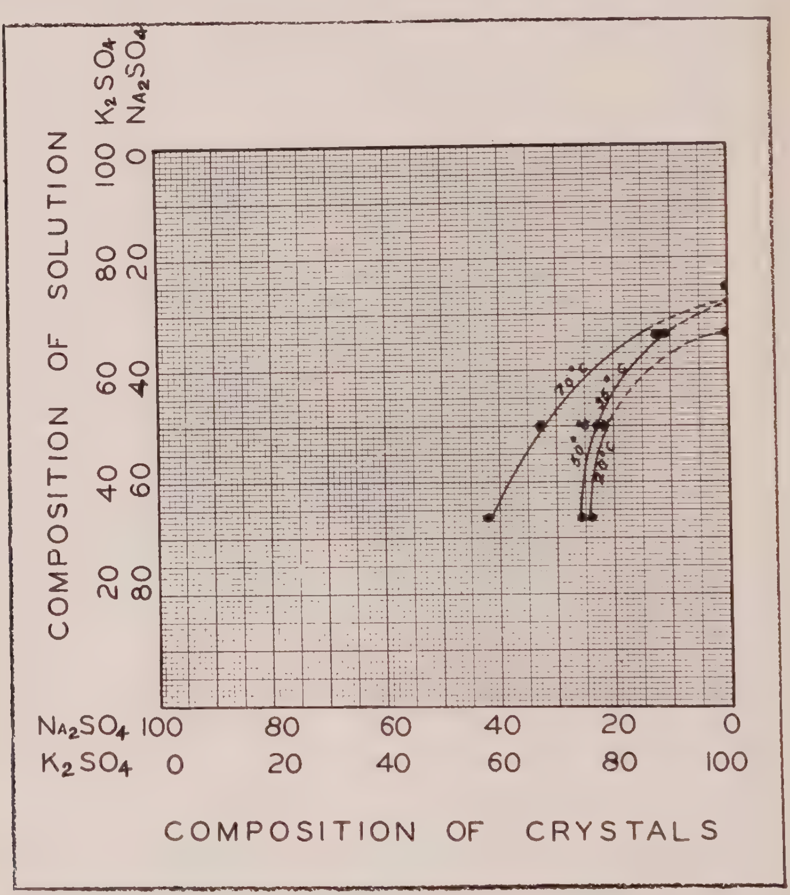

Fig. 2

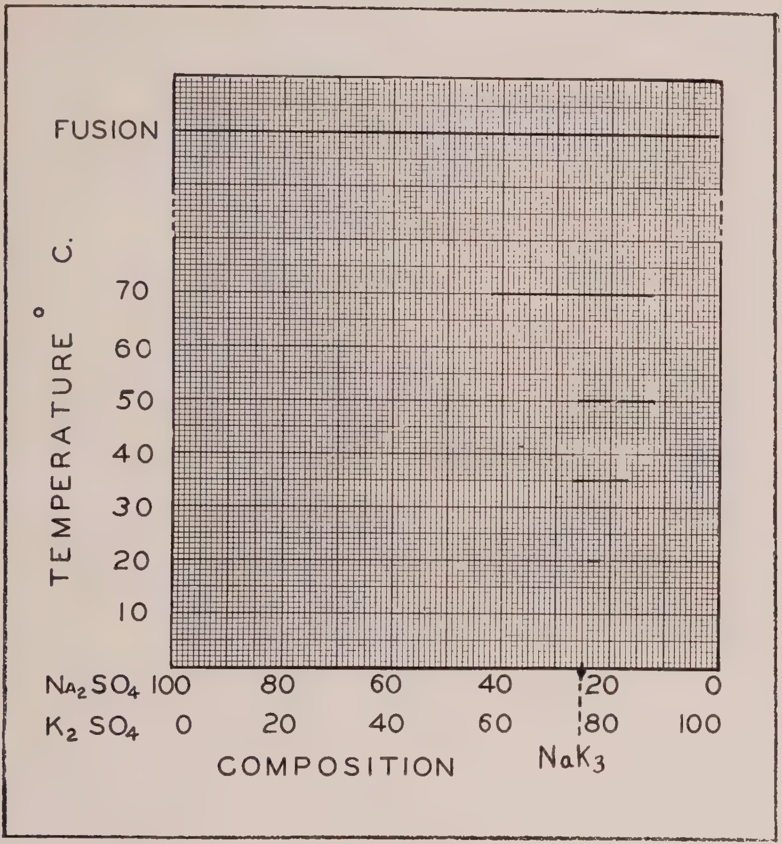

Fig. 3

The Crystal Structure of the Isomorphous Series Na_2SO_4 *I, Aphthitalite, and* β-K_2SO_4

Gossner (1928) described a structure for $NaK_3(SO_4)_2$, and Bellanca (1943) reported a closely related structure for $NaKSO_4$. Bredig (1941) proposed a structure type A_2XO_4, with a hexagonal unit cell $P\bar{3}m1$ and $Z=2$, for a series of compounds, principally at high temperatures. These included $CaSiO_4$ and alkaline earth phosphates, as well as alkali sulfates. No atomic positions were suggested.

In the space group $P\bar{3}m1$, the positions of the 4 metal atoms are not all structurally equivalent. Two occur at $\pm\frac{1}{3}\,\frac{2}{3}\,z$; one at 000 and one at 00 $\frac{1}{2}$. In these three types of positions the number and arrangement of the oxygen atoms surrounding the metal atoms are different. For the structures of $NaK_3(SO_4)_2$ and $NaKSO_4$, as determined by Gossner and Bellanca, respectively, the distances from the metal to oxygen vary considerably. In all cases they are greater than the sum of the commonly used values of the ionic radii, and for one position considerably greater. This could account for the unusually small changes in unit cell dimensions with marked changes in the relative proportions of Na and K, and also offers an explanation of why these two elements, which do not usually replace each other in solid solution series, do so in this case.

It would seem most probable that the replacement of Na by K is selective, rather than occurring randomly over the 2+1+1 metal positions. If, starting with a low K content, additional K ions can replace Na ions in positions where the Na to O distance is greater than necessary (in so far as required by Na radius+O radius), the impact on unit cell dimensions will not be appreciable until these positions are filled. This would explain the comparatively small changes in the values of a_o and c_o until outside the 4:1 and 1:4 range of Na and K. The comparatively large change in c_o at the K end of the series occurs when even the positions with least room, originally filled by Na, must now be occupied by K. The smaller change, chiefly in a_o, at the Na end of the series may be attributed to the fact that here the cell dimensions are chiefly controlled by the close packing of O atoms.

The Relation Between Lithium Sulfate and Potassium Sulfate

The System Li_2SO_4-K_2SO_4-H_2O (*Aqueous solutions*)

Li_2SO_4 is not known to occur as a natural mineral. It is monoclinic and may be prepared by fusion of the common reagent salt, $Li_2SO_4 \cdot H_2O$, which is also monoclinic. The ionic radii of Li^+ and K^+ are 0.78 Å and 1.33 Å, respectively. The earliest investigation of the compound $LiKSO_4$ goes back to Schabus (1855) who erroneously described it as an isomorphous mixture of Li_2SO_4 and K_2SO_4. Later the substance was investigated by Rammelsberg (1866), Scacchi (1867), Wyrouboff (1880, 1882 and 1890), Wulff (1890, 1893), and Traube (1892 and 1894). Their descriptions were concerned with the morphology of the crystals which were found to be hexagonal pyramidal, with $a:c=1:1.6755$ (Traube, 1892).

During the crystallization of mixtures of lithium sulfate and potassium sulfate from aqueous solutions, the common reagent salt $Li_2SO_4 \cdot H_2O$ was used. Two types of solutions were prepared, namely, neutral (i.e. the pure salts dissolved in distilled water) and acidic (i.e. a few drops of concentrated H_2SO_4 were added to the solution in distilled water). Crystallization was made at room temperature and at 70° C. (±3°). The compositions of the solutions were in the following molecular ratios when calculated in terms of Li_2SO_4:K_2SO_4=4.30:1, 3.44:1, 2.58:1, 1.72:1, 0.86:1, 1:2.32, and 1:3.48, respectively. Platy hexagonal crystals, with well developed basal pinacoids, were formed from most of the solutions. Laue photographs show hexagonal symmetry, and Weissenberg photographs yielded the space group $P6_3$, with $a_o = 5.13$Å and $c_o = 8.60$ (±0.02Å), in agreement with the data of Bradley (1925) for $LiKSO_4$. Powder photographs of crystals grown from the various solutions at two different temperatures were made, with the results shown in Table 4.

TABLE 4

Mol. ratio of Li_2SO_4:K_2SO_4 in solution	Type of x-ray powder photographs of crystals formed			
	at 70° C.		at 20° C.	
	From Neut. sol.	Acid sol.	Neut. sol.	Acid sol.
4.30:1.0	$LiKSO_4$	mixture of $Li_2SO_4 \cdot H_2O$ and $LiKSO_4$	$LiKSO_4$	$LiKSO_4$
3.44:1.0	$LiKSO_4$	$LiKSO_4$	$LiKSO_4$	$LiKSO_4$
2.58:1.0	$LiKSO_4$	$LiKSO_4$	$LiKSO_4$	$LiKSO_4$
1.72:1.0	$LiKSO_4$	$LiKSO_4$	$LiKSO_4$	$LiKSO_4$
0.86:1.0	$LiKSO_4$	$LiKSO_4$	β-K_2SO_4	β-K_2SO_4
1.00:2.32	mixture of β-K_2SO_4 & $LiKSO_4$	$LiKSO_4$	β-K_2SO_4	β-K_2SO_4
1.00:3.48	β-K_2SO_4	β-K_2SO_4	β-K_2SO_4	β-K_2SO_4

The powder pattern data are given in Table 5, There is no change in the spacings of the lines on the films, indicating that the crystals represent a compound of fixed chemical composition, with no solid solution relationship. As an additional check, chemical analyses were made. These are shown in Table 6, and the results show clearly that the potassium content in crystals grown from different solutions is almost constant, and corresponds to the 1:1 ratio of K:Li.

The System Li_2SO_4-K_2SO_4 (*Fused samples*)

Powder photographs of samples prepared by fusing mixtures of Li_2SO_4 and K_2SO_4 in various proportions likewise indicate clearly that there is no solid solution relationship. Only one mixture of the approximate ratio of 1:1 gave rise to the $LiKSO_4$ pattern alone. All other mixtures gave the $LiKSO_4$ pattern together with that of either Li_2SO_4 or K_2SO_4, depending upon the proportions of the mixture. Thus even at the higher

TABLE 5. POWDER PHOTOGRAPH DATA FOR $LiKSO_4$

d observed	d calc.	$hk \cdot l$	I observed
4.33 Å	4.31 Å	00 2	1
4.00	3.96	10·1	10
3.12	3.10	10·2	9
2.58	2.57	11·0	7
2.47	2.47	11·1	2
2.22	2.225 2.21	20·0 11·2	5
2.16	2.165 2.155	20·1 00·4	5
1.985	1.98	20·2	1
1.66	1.655 1.650	21·1 11·4	3
1.61	1.61	10·5	1
1.58	1.57	21·2	3
1.50	1.49	30·0	3
1.37	1.36	20·5	2

TABLE 6

Composition of solution in terms of mol. ratio $Li_2SO_4:K_2SO_4$	Percentage of K in crystals	Theoretical percentage of K in $LiKSO_4$	Type of powder pattern
3.44:1	27.8	27.5	$LiKSO_4$
2.58:1	27.0	27.5	$LiKSO_4$
1.72:1	27.7	27.5	$LiKSO_4$
0.86:1	28.6	27.5	$LiKSO_4$
3.44:1*	28.0	27.5	$LiKSO_4$
2.58:1*	28.2	27.5	$LiKSO_4$

* Crystallized from slightly acid solution.

temperature near fusion, the $LiKSO_4$ structure does not permit substitution of either Li or K for the other, unless there was unmixing during cooling. The cooling was relatively rapid, and if there was any tendency for a solid solution to form, it is probable that it would have persisted as a metastable phase, especially close to the 1:1 ratio.

THE RELATION BETWEEN LITHIUM SULFATE AND SODIUM SULFATE

The System Li_2SO_4-Na_2SO_4-H_2O (*Aqueous solutions*)

The earliest investigation of crystals of $LiNaSO_4$ found reported is that of Scacchi (1867), who determined the axial ratio for the ditrigonal pyramidal crystals to be $a:c=1:0.5624$. Traube (1892) reported the specific gravity of $LiNaSO_4$ as 2.369. This last value was not confirmed in the present study. The crystals used by these early investigators were obtained from aqueous solutions at about 50° C., and also from acid solutions, with or without the addition of NaCl.

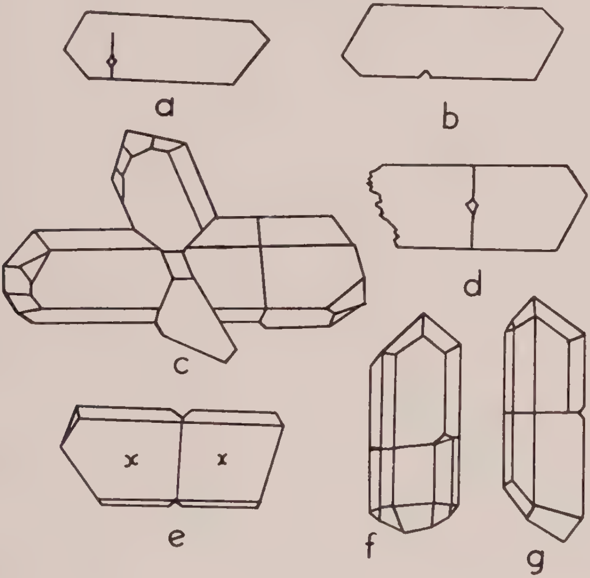

FIG. 4

Crystals were grown from solutions containing a wide range of ratios of Li_2SO_4 and Na_2SO_4. Better crystals were obtained when a few drops of H_2SO_4 were added to the solutions, and crystallization was carried out at about 70° C. Most of the crystals were elongated parallel to the *c* axis. A fairly common feature observed on these crystals is an equatorial marking around the prism zone. Figure 4 shows some free hand drawings of typical crystals as they appear under low magnification with the binoculars. In most cases the mark appears as an even straight line, which may completely surround the crystal (Fig. 4, *c*, *d*, *e*, *f* and *g*), or only partly surround it (Fig. 4*a*). Under higher magnification, this line shows as a series of tiny disconnected re-entrant angles. In many cases at the intersection of this line with an edge, there is a more prominent re-entrant angle (Fig. 4, *b*, *e* and *g*). On the two circle goniometer, the parts of a prism face on either side of the line either reflect light simultaneously, or else give signals separated by only a few minutes of arc. Usually the prism faces continue uninterrupted on both sides of the line (Fig. 4, *d* and *e*), but in some cases the faces are interrupted, and different forms are shown on the two sides (Fig. 4, *c*, *f*, and *g*). Since the crystals of $LiNaSO_4$ do not have a horizontal symmetry plane, it is believed that this line represents twinning on (0001).

No previous *x*-ray work has been reported on $LiNaSO_4$. Laue photographs at first glance appear to show a 6-fold axis of symmetry, but careful inspection reveals a few reflections which indicate the true symmetry to be 3-fold. (Laue symmetry $\bar{3}\ 2/m$). This is verified by Weissenberg photographs. Lattice constants of the hexagonal unit cell (*P*) were found to be $a_o = 7.64$Å and $c_o = 9.76$Å (± 0.02Å), giving an axial ratio of $a:c = 1:1.277$. The pyramidal faces gave poor reflections. Measurements on the two circle goniometer show two sets of pyramidal faces, separated by 30°, which in terms of the unit cell dimensions are 10·2 and 11·4. If the latter were assumed to be the unit pyramid 10·1, the derived axial ratio would be 0.58, close to that given by Scacchi. The characteristic missing reflections on the Weissenberg photographs are $hh\bar{2h}l$ with l odd, indicating a *c* glide plane. In the Laue class $\bar{3}\ 2/m$ the only possible space groups with a *c* glide are $P\bar{3}1c$ and $P31c$. Since $LiNaSO_4$ crystals are known to be pyroelectric (Traube, 1892), space group $P\bar{3}1c$ is ruled out.

The specific gravity of the crystals was determined by diluting acetylene tetrabromide until they remained suspended in it. The specific gravity of the liquid was then found to be 2.515, which is quite different from the value of 2.369 reported by Traube (1892). Because of this discrepancy, an approximate check was made with the Gladstone and Dale Law. Using the specific refractive energy values (Larsen and Berman, 1934) of $LiNaSO_4$, and the refractive indices, namely $\epsilon = 1.495$ and $\omega = 1.490$, the specific gravity of this compound was determined to be 2.54. Using the experimentally determined value of 2.515, the number of formula weights of $NaLiSO_4$ in the unit cell was found to be $5.97 \cong 6$.

Powder photograph data are given for $LiNaSO_4$ in Table 7. No change in the spacings of the powder patterns of the various samples was found, indicating the absence of any solid solution relationship between Li_2SO_4 and Na_2SO_4. Since these two compounds and the compound $LiNaSO_4$ all crystallize in different systems, no extensive solid solution would be expected.

The System Li_2SO_4-Na_2SO_4 (*Fused samples*)

X-ray powder photographs were made of samples prepared by fusing mixtures of varying proportions of Li_2SO_4 and Na_2SO_4. Only one mixture of the approximate ratio 1:1 gave rise to a pattern consisting solely of

TABLE 7. POWDER PHOTOGRAPH DATA FOR $LiNaSO_4$

d observed	*d* calc.	*hk·l*	*I* observed
3.94 Å	3.95 Å	10·2	8
3.83	3.82	11·0	10
3.04	3.02	11·2	7
2.94	2.94	10·3	7
2.75	2.745	20·2	9
2.425	2.445	00·4	4
	2.430	12·1	
2.31	2.32	20·3	3
2.215	2.23	12·2	3
	2.20	30·0	
2.15	2.15	30·1	4
2.01	2.01	30·2	1
1.97	1.97	20·4	2
1.91	1.91	22·0	5
1.88	1.88	10·5	1
1.83	1.83	13·0	2
	1.825	30·3	
1.745	1.75	12·4	1
1.69	1.685	20·5	1
1.64	1.65	40·0	2
	1.635	40·1	
	1.63	00·6	
1.60			2
1.565			2
1.545			2
1.505	1.50	11·6	3
1.47			4
1.44			4

$LiNaSO_4$ lines. All other ratios gave the pattern of $LiNaSO_4$ together with that of either Li_2SO_4 or Na_2SO_4, thus verifying the results obtained from the crystals. As was mentioned in the case of fused Li-K sulfate mixtures, there is the possibility of limited solid solution near the fusion temperature, with unmixing on cooling.

The System $(NH_4)_2SO_4$-K_2SO_4-H_2O (*Aqueous solutions*)

Ammonium sulfate occurs in nature as the mineral mascagnite. Both mascagnite and arcanite (β-K_2SO_4) are orthorhombic and have the space group *Pmcn* (Winchell, 1951). Because of the isomorphism of the two compounds and the similarity of the ionic radii of NH_4 and K (1.43 and 1.33Å, respectively) complete solid solution between the two would be expected. Such was reported by Fock (1892). *X*-ray evidence completely substantiates this. Crystals formed from solutions of varying proportions do not have the same composition as the solution (Table 8), but are shown by both Weissenberg and powder photographs to be structurally alike, with gradual decrease in lattice constants with increasing potassium.

	a_o	b_o	c_o
$(NH_4)_2SO_4$	5.95 Å	10.56 Å	7.72 Å
NH_4KSO_4	5.84	10.08	7.52
β-K_2SO_4	5.70	9.99	7.34

The measured spacings and intensities of the powder photograph lines of various NH_4-K sulfate solid solutions are in agreement with the published data for the mineral taylorite as given by Winchell and Benoit (1951).

The crystals obtained from the various solutions of mixtures of potassium and ammonium sulfates were chemically analyzed so as to determine potassium quantitatively. A known amount of the dry powdered material was ignited first to remove NH_3 and the amount of potassium was then determined by the periodate method (Willard and Boyle, 1941). The results of the chemical analysis are given in Table 8. The table shows clearly how the composition of the crystals varies continuously with the change in the composition of the solution.

TABLE 8

Comp. of solution $K_2SO_4:(NH_4)_2SO_4$	Temp. of crystallization	% K determined quant.	Type of x-ray powder pattern	Comp. of crystals $K_2SO_4:(NH_4)_2SO_4$
25.0:75.0	70° C.	21.05	xtalline solid soln.	46.3:53.7
33.3:66.7	70° C.	31.8	xtalline solid soln.	70.0:30.0
50.0:50.0	70° C.	39.4	xtalline solid soln.	86.5:13.5
66.7:33.3	70° C.	42.8	xtalline solid soln.	94.0: 6.0
75.0:25.0	70° C.	43.2	xtalline solid soln.	95.0: 5.0

THE RELATION BETWEEN SODIUM SULFATE AND AMMONIUM SULFATE

The System Na_2SO_4-$(NH_4)_2SO_4$-H_2O (*Aqueous solutions*)

No mineral having the composition of pure sodium ammonium sulfate has been reported. Ammonian aphthitalite, $(KNH_4)_3Na(SO_4)_2$, from the Chincha Islands has been described by Frondel (1950). Although Na_2SO_4 III, Na_2SO_4 V and $(NH_4)_2SO_4$ are all orthorhombic, their space groups are different. Since the ionic radii of NH_4 and Na are quite different, no solid solution of the $(NH_4)_2SO_4$ with either form of Na_2SO_4 would be expected. This was verified by powder photographs of crystals obtained from solutions of varying proportions of the two sulfates at 70° C. No evidence of solid solution was found. Samples could not be prepared by melting, since ammonium sulfate dissociates readily at higher temperatures.

SUMMARY AND CONCLUSIONS

1. Aphthitalite, a sodium potassium sulfate mineral, has been prepared in the laboratory from aqueous solutions as well as from fused melts of various sodium and potassium sulfate proportions. Using x-ray methods, powder, rotation and Weissenberg, it is revealed that at high temperatures close to fusion, sodium sulfate and potassium sulfate are isomorphous and form a complete series of solid solutions. The same synthetic mineral, prepared from aqueous solution has been found, by x-ray investigation and checked by chemical analysis, to have a limited range of solid solution varying between the ratios of 1K:1Na to 5K:1Na at 70° C. and is limited to the almost invariable ratio of 3K:1Na at room temperature. A gradual increase in the lattice constants as well as in the refractive indices has been observed, with increasing potassium content in the synthesized aphthitalites of the high-temperature complete solid solution series. It is concluded, therefore, that aphthitalite whether of composition $NaK_3(SO_4)_2$ or $NaKSO_4$ is to be considered simply as a solid solution member of the high temperature forms of sodium sulfate and potassium sulfate.

2. $LiKSO_4$ and $LiNaSO_4$, both hexagonal but with different space groups (i.e. different structures), are found to represent double compounds between Li_2SO_4 and K_2SO_4 and Li_2SO_4 and Na_2SO_4, respectively. No solid solution relationship was revealed by x-ray analysis.

3. The following data have been determined for $LiNaSO_4$: $a_o = 7.64$, $c_o = 9.76$ (± 0.02Å); $a_o : c_o = 1:1.277$; $Z = 6$; space group $P31c$; measured specific gravity 2.515, calculated specific gravity 2.527. Twinning on (0001) common.

4. Complete solid solution and isomorphous relationship between ammonium sulfate and potassium sulfate have been verified by x-ray methods.

5. There is no solid solution between sodium sulfate and ammonium sulfate at 70° C.

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NOTES AND NEWS

ORIENTED OLIVINE INCLUSIONS IN DIAMOND*

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According to Sutton (1) and Williams (2) there has been strong evidence for the presence of olivine inclusions in the diamond. These authors, however, indicate that there was not definite proof of olivine being completely enclosed within a diamond.

Recently a number of diamond crystals with interesting inclusions were observed at this laboratory. These were noticed during an investigation of the double refraction of several thousand diamonds of the type known as *flats*. These inclusions are colorless and, in most cases, well developed. They are oriented parallel to the (111) plane and elongated parallel to the [101] direction of the diamond. They exhibit inclined extinction. Due to the orientation of these inclusions within the diamond, it seemed feasible to establish their identity by *x*-ray analysis.

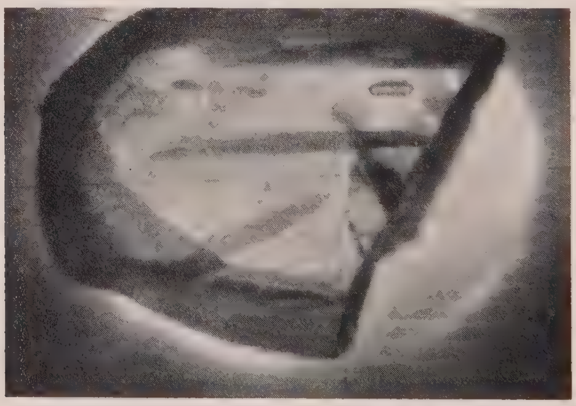

FIG. 1. Diamond *flat* containing three parallel olivine inclusions, ×28. Only two of these inclusions are in focus.

Using the direction of elongation of the inclusions (diamond [101]) as the rotation axis, zero and first level Weissenberg, and rotation photographs were made of one of the specimens containing three parallel inclusions (Fig. 1). Since the largest inclusion is isolated at one corner of the *flat*, it was possible to obtain *x*-ray patterns from this single inclusion superimposed upon diamond patterns. A study of these films definitely established the identity of this inclusion as being olivine elongated along its [101] direction. A second diamond containing a similar inclusion was subsequently investigated in the same manner. This inclusion also proved to be olivine with the same elongation.

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* Contribution from the Department of Mineralogy and Petrography, University of Michigan, No. 177.

After the identity of these inclusions was established by x-ray methods, an optical check verified this peculiar elongation of olivine and also explained the inclined extinction that was observed prior to the identification of the inclusions. In Fig. 2, lines a and c correspond to these crystallographic axes which are also the extinction directions. The b axis is perpendicular to the (111) face of the *flat* which is parallel to the plane of the photograph. The extinction angles measured against the direction of elongation of the inclusion correspond to the calculated angles between the [101] direction and the a and c axes of olivine. The observed extinction was somewhat wavy, perhaps due to strain.

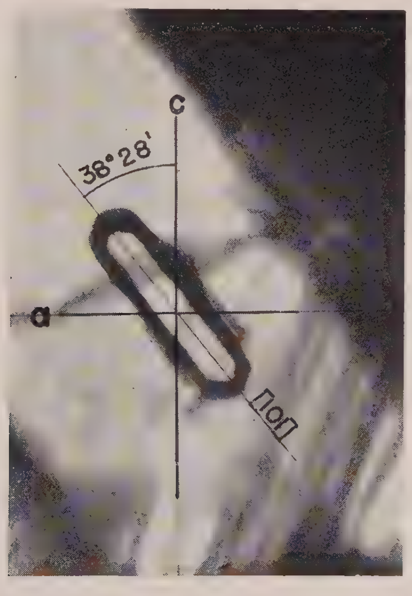

Fig. 2. The largest olivine inclusion of Fig. 1. The a and c crystallographic axes and elongation direction [101] are indicated.

Inclusions similar to those described above were noted in a number of other diamond *flats* which were examined. A detailed investigation, however, has not been carried out.

This work was incidental to the general investigation of diamond which is being carried on with the financial assistance of the Office of Naval Research. The writers wish to thank Professor L. S. Ramsdell and Professor C. B. Slawson, of the Mineralogical Laboratory, University of Michigan, for their helpful criticisms and suggestions. The writers would also like to thank Mr. Charles Koebel, of Detroit, who furnished the diamonds used in this investigation, and presented those with unusual features to the University collections.

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NOTES ON SOME MINERAL OCCURRENCES IN THE GUFFEY REGION, COLORADO*

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Introduction

Most of these mineral occurrences have not been described previously in detail, although a few of them have been reported briefly. The mineral localities here described were studied during the summers of 1950 and 1951 as a part of field investigations leading to the preparation of a doctoral dissertation in mineralogy for the University of Michigan.

The Guffey region is an area of about 125 square miles in north-central Fremont County and southeast Park County in the Front Range of central Colorado. Its center is 34 miles by road northwest of Canon City. The area is underlain chiefly by pre-Cambrian igneous and metamorphic rocks, which have been covered to the north, east and west by post-Laramide extrusive rocks.

On the basis of their geological environments it is possible to classify the mineral occurrences under three headings: pegmatite minerals, minerals of the metamorphic rocks and minerals of metallic ore deposits.

Pegmatite Mineral Occurrences

The pegmatites in the area are pre-Cambrian in age, and probably are associated genetically with the intrusion of the Pikes Peak granite batholith (Bever, 1952).

Euxenite: An unusual concentration of this rare radioactive mineral was found in the core margin unit of a small zoned pegmatite about 200 feet west of the Mac Gulch drainage, approximately 3 miles southeast of Guffey. The euxenite occurs in bladed crystals as much as 2 centimeters in length, whose freshly broken surfaces are glassy and brownish-black in color. Associated are abundant coarse biotite and magnetite as well as the other common granitic pegmatite minerals. Tantalite also is present in relative abundance. A few grains of altered euxenite also were found at the East-West pegmatite which is described below.

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* Contribution from the Department of Mineralogy and Petrography, University of Michigan, No. 178.

Monazite: A number of small crystals and crystal fragments of monazite were found at the East-West feldspar pegmatite, which is about 1 mile west-northwest of the Meyers Ranch pegmatite described by Hanley *et al.* (1950, pp. 105–107). Associated with the monazite in addition to the common pegmatite minerals are beryl, schorl, tantalite, and smoky quartz.

Magnetite-quartz-sillimanite selvage: The Whopper pegmatite, which is in the Micanite group, has been described by Hanley *et al.* (1950, pp. 48–50) who report "granular magnetite masses as much as 3 inches in size and a mineral tentatively identified as sillimanite." Pods of coarse-grained magnetite-quartz-sillimanite rock, as much as 25 centimeters in diameter, occur as a discontinuous selvage along the pegmatite margins. Individual magnetite octahedra are as large as 3 centimeters in diameter. In some pods the magnetite and quartz are intergrown simplectically to graphically. Similar pods of magnetite-quartz-sillimanite rock were observed along or near the margins of a number of other pegmatites in the Guffey area.

MINERAL OCCURRENCES IN METAMORPHIC ROCKS

The pre-Cambrian country rocks include those which have been correlated with the Idaho Springs formation and closely associated migmatite.

Cordierite: Sterrett (1923) recognized cordierite in the Climax pegmatite, which is one of the main deposits of the Micanite group. Hanley *et al.* described this deposit (1950, pp. 43–45) and Heinrich (1950) discussed the cordierite occurrence in detail, including its extensive alteration to pinite and summarized and evaluated the various theories concerning the origin of pegmatitic cordierite. Large boulders and several outcrops of cordierite-sillmanite gneiss were found 300 feet west of the Climax pegmatite. This rock contains metacrysts of glassy gray-violet cordierite as much as 3 millimeters in diameter. This occurrence, previously unknown, undoubtedly must be considered in re-evaluating theories on origin of the nearby pegmatitic cordierite.

Eckel in 1932 (Lovering and Goddard, 1950) found "a little cordierite (?)" in the ore-bearing zone at the Lone Chimney mine, now known as the Betty mine. This occurrence was confirmed by the author, but more noteworthy is the occurrence near the margin of the same mineralized zone about two-thirds of a mile south of the mine on the north bank of Thirty-one Mile Creek. The outcrop is about 500 feet east of the Copper King prospect described by Hanley *et al.* (1950, p. 105). Cordierite and quartz in granular aggregates as much as 4 centimeters across are interstitial to radiating fibers of anthophyllite which may reach a length of 3 centimeters. This cordierite is the rarer optically positive variety.

Kyanite: Blades of kyanite, 5×2 centimeters, partly or completely altered to fibrous sillimanite occur randomly oriented in injection gneiss on the north bank of Dicks Gulch about 2 miles west of Currant Creek. The other minerals of the gneiss are quartz, muscovite, biotite, sillimanite, microcline and plagioclase. Kyanite has previously not been reported from this part of Colorado. Aggregates of sillimanite needles in gneisses in other parts of the area also appear, because of their shape, to be pseudomorphous after single kyanite crystals.

Sillimanite: Previous reports on the Guffey area mention the presence of sillimanite, but do not indicate that it is unusually abundant. Sillimanite is almost invariably present in large quantities in transitional zones of injection gneiss that lie between schists of the Idaho Springs formation and the Pikes Peak granite pluton. At the junction of Currant Creek and Lues Gulch just a few feet north of Colorado State Highway 9 is a lens of relatively pure sillimanite-quartz rock with accessory biotite, feldspars and sericite. This deposit is approximately 175 feet long and as much as 30 feet wide, concordant with the foliation of the flanking injection gneiss and localized along the gneiss-Pikes Peak granite contact. This sillimanite is white to greenish white in color, although at the surface much of it is heavily stained by iron oxides.

An erosion surface of several square miles, ½ to 2 miles east of Micanite Ridge, is underlain chiefly by sillimanite schist containing a high percentage of magnetite. Outcrops of this rock generally weather dark reddish-brown to black.

Minerals of Metallic Ore Deposits

Scheelite: Tungsten mineralization has heretofore not been reported from this area, but scheelite occurs as disseminated grains and crystals in possible commercial concentrations along the ridge about one-quarter mile northeast of the West ranch, which is at the junction of Colorado State Highway 9 and the Guffey road. The scheelite occurs in a coarse-grained crystalloblastic aggregate of epidote, hornblende, quartz, calcite, and garnet probably formed by the metamorphism and metasomatism of an impure calcareous sediment. The scheelite is milky white, and some of the crystals are unusual in showing a pronounced zonal variation in fluorescent colors. The centers of the crystals fluoresce blue-white, whereas the narrow margins exhibit a yellow fluorescent color. In view of the studies by Cannon *et al.* (1942) and by Greenwood (1943) this indicates a core of tungsten-rich material and a molybdenum-rich margin. Grains of scheelite occur rarely in other similar rocks throughout the area.

Minerals at the Betty mine: The Betty mine was examined by E. B.

Eckels in 1932 and his description is included in the report of Lovering and Goddard (1950). The mine is about one mile west of Currant Creek, one-third mile south of Black Mountain road. A 300-foot shaft has been sunk near the junction of a rhyolite breccia dike and a diabase dike, both of which have been intruded into injection gneiss.

In addition to malachite, azurite, galena, and chalcopyrite reported by Eckels the writer found significant amounts of pyrite, bornite, and sphalerite and minor amounts of covellite, either in hand specimens or in polished sections. Some of the chalcopyrite has formed as plates along the cleavages of amphibole crystals.

Although vesuvianite reported by Eckels was not confirmed, abundant green spinel is present as small disseminated grains that impart a greenish cast to the quartz-calcite-garnet-amphibole rock in which it occurs. Some octahedra of spinel attain a size of approximately 2 centimeters on edge.

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NOTE ON SPHALERITE AND WURTZITE*

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Interest in the crystal forms of zinc sulfide was aroused by its similarity to SiC, whose various structures have been investigated by members of the Mineralogy Department of the University of Michigan. Commonly natural wurtzite occurs in the form referred to as $2H$ by the Ramsdell (1947) nomenclature. In addition natural wurtzite was found by Frondel and Palache (1950) to occur in the forms $4H$, $6H$, and $15R$[1]. An investigation was undertaken to see if synthetic wurtzite could be crystallized from aqueous solution in these and in the other forms that have been identified for SiC.

Allen and Crenshaw (1914) reported the conditions under which the various forms of ZnS were precipitated. They identified sphalerite, wurtzite, and amorphous ZnS by optical methods. At temperatures below 250° C. the precipitate was so fine-grained that they called the material "amorphous." However by *x*-ray methods it is easy to establish that this apparently amorphous material is crystalline and also to determine its structural modification. At the time of these early investigations, *x*-ray techniques were not in general use so it seemed advisable to repeat these experiments, using *x*-ray powder photographs to identify the precipitates.

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* Contribution from the Department of Mineralogy and Petrography, University of Michigan, No. 179.

[1] Müller (1952) has recently succeeded in producing these same modifications by tempering synthetic ZnS crystals up to 10 hours at 870–905° C.

ZnS was precipitated at temperatures ranging from 15° C. to 100° C. from strongly alkaline to acidic zinc sulfate solutions by adding hydrogen sulfide, sodium sulfide, or ammonium sulfide. In all cases the precipitates had the sphalerite structure. In order to obtain precipitates above 100° C., the closed double tube method of Allen and Crenshaw was used. These workers claimed to have obtained 100% wurtzite by precipitating ZnS from a zinc sulfate solution containing 4% sulfuric acid by weight at 250° C. with a reaction time of 48 hours. Their experimental procedure was repeated and our product was identified as 100% sphalerite by means of *x*-ray powder photographs. In several succeeding attempts, the initial acid concentration was varied, and reactions were also carried on in alkaline solutions. The results were always the same: the product was sphalerite.

X-ray examinations showed that reagent quality ZnS[2] is predominantly wurtzite with small amounts of sphalerite. The manufacturer stated (written communication) that the ZnS was precipitated as sphalerite and roasted in an inert atmosphere in order to volatilize impurities. Fuller (1929) has prepared 100% wurtzite by heating sphalerite with NaCl at 1100° C. until the NaCl was completely volatilized. The manufacturer of the reagent ZnS realized that they were converting the sphalerite to wurtzite but made no attempt to insure complete conversion. We heated this ZnS at 250° C. with sulfuric acid of various concentrations similar to those used by Allen and Crenshaw in glass tubes sealed in steel bombs. Under these conditions all of the wurtzite was reconverted to sphalerite in 48 hours.

These results cast doubt upon the validity of the conclusions of Allen and Crenshaw which have been commonly used as a basis for determining the genetic relations in ores containing wurtzite.

The writer wishes to thank Professor C. B. Slawson of the University of Michigan for his kind cooperation in this project, which was carried on with the financial assistance of the Office of Naval Research.

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[2] Baker and Adamson Co., New York.

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Symposium on Quartz Oscillator Plates

This popular number of the *American Mineralogist* for May–June, 1945, has been reprinted for the third time. Fourteen authoritative articles in 264 pages, illustrated. Price $3.00, postpaid. Send orders to the Treasurer, Dr. Earl Ingerson, U. S. Geological Survey, Washington 25, D. C.

Canadian Report Published

The Second Annual Report, 1951–52, *National Advisory Committee on Research in the Geological Sciences*, W. A. Bell, Chairman, was published, October 1952, by the Geological Survey of Canada, Department of Mines and Technical Surveys, Ottawa, Ontario, Canada. The Report is mimeographed on legal sized sheets; included in its 87 pages is an Appendix listing conditions governing award of grants for research to Canadian Universities, a list of grants awarded for 1951–52, an 8-page section on "The Year in Review," and Reports of eight Subcommittees.

AGI Officers for 1952–53

Carey Croneis and Harry S. Ladd were unanimously re-elected as President and Secretary-Treasurer, respectively, at the recent meeting of the Board of Directors in Boston, Massachusetts.

Vice-President for the coming year is W. C. Krumbein, geology professor at Northwestern University and AGI Board member representing SEPM.

Professor Paul Niggli, one of the Correspondents of the Mineralogical Society of America and recipient of the Roebling Medal in 1947, died of a heart attack Jan. 13, 1953, at the age of 64.

O. Ivan Lee, research spectrographer for the Crocker Cancer Institute of Columbia University and Chief spectrographer of the United States Testing Co., died on Nov. 25, 1952, at the Medical Center, Jersey City, New Jersey, at the age of 64. He was one of the founders of the New Jersey Mineralogical Society.

At the Tenth Honor Awards Convocation held on Dec. 16, 1952, in the Interior Building, Washington, D. C., Dr. Waldemar T. Schaller received the Distinguished Service Gold Medal Award of the Department of the Interior, in recognition of an eminent career in the field of science. During his 47 years of service on the Geological Survey Dr. Schaller has published 165 papers dealing with every phase of mineralogy, including the description and naming of 43 new minerals.

BOOK REVIEWS

TABELLEN ZUR OPTISCHEN BESTIMMUNG DER GESTEINSBILDENDEN MINERALE, BY W. E. TRÖGER. xi+147 pp., 17 tables, 376 figures, 2 plates in rear pocket. E. Schweizerbart'sche Verlagsbuchhandlung, Stuttgart, Germany. DM. 27.80. ($6.67) 1952.

Professor Tröger, of the Mineralogical Institute of the Technical University of Darmstadt, has succeeded in providing workers in optical mineralogy and microscopic petrography with a highly useful and effective tool in his compilation, Tables for the optical determination of the rock forming minerals. The work is essentially without formal text, except for the introduction and an explanation of abbreviations. Most of its data are summarized in 6 tables:

1. Water soluble minerals.
2. Opaque or nearly opaque minerals.
3. Uniaxial or pseudocubic minerals.
4. Uniaxial tetragonal minerals.
5. Uniaxial hexagonal minerals.
6. Biaxial minerals.

The last are arranged chemically under oxides, hydroxides, carbonates, sulfates, phosphates, nesosilicates, sorosilicates, inosilicates, phyllosilicates and tectosilicates. Each table gives information under the headings: (1) Name, system, chemical composition; (2) Common forms, twinning, habit, aggregate structure; (3) Cleavage, gliding, hardness, density; (4) Optical orientation; (5) Refringence and birefringence; (6) Angle and dispersion of the optic axes; (7) Color in thin section, pleochroism, absorption; (8) Special diagnostic chemical characteristics; (9) Diagnostic characteristics of similar minerals; and (10) Paragenesis. An amazing amount of information has been packed into the tables, although the descriptions of the parageneses are necessarily curtailed and thus somewhat oversimplified.

With each of the tables are numerous figures of two types: (1) Three dimensional and sectional crystal drawings showing the optic orientation and (2) Curves relating compositional variation to variations in optical properties and density. The orientation diagrams are unusually good and are alone worth the price of the book. Variation diagrams are given for garnets, spinels, opal, melilite, vesuvianite, scapolite, rhombohedral carbonates, tourmalines, beryl, cancrinite, davyn, columbite-tantalite, amblygonite-montebrasite, topaz, andalusite, staurolite, epidote, piedmontite, allanite, axinite, cordierite, prehnite, rhodonite-wollastonite, orthopyroxenes, augitic pyroxenes, aegirine, jadeite, anthophyllite, cummingtonite, actinolite, glaucophane, arfvedsonite, hastingsite, hornblende, muscovite, biotite, lepidolites, margarite, chlorites, stilpnomelane, montmorillonite, orthoclase, plagioclase (19 curves) and thomsonite. Unfortunately most of the curves are expressed in molecular percentage of end-member molecules, even for systems of great complexity. The majority of the curves are either new ones constructed by Tröger or are taken from Winchell's fourth edition of Elements of Optical Mineralogy. The pyroxene diagrams are mainly those of Hess and the plagioclase ones those of Köhler and Nieuwenkamp. In many cases, e.g. the epidote group, cordierite, sodic amphiboles, biotite, lithium micas, margarite, and chlorites, the diagrams are sharply oversimplified or are constructed from optical-chemical information now known to be archaic. Must we force extreme extrapolations for minerals for which our data are admittedly insecure? Were it not better to admit gaps in our knowledge than to perpetuate misconceptions? Compilations are valuable to the extent that they are critical.

Also in the work are a number of diagrams relating to the determination and interrelation of optical constants, and finally, "finding" tables based on density, dispersion, optic figures on cleavage pieces, color and pleochroism and miscellaneous observations.

No advanced mineralogist will wish to be without this book, which will undoubtedly have widespread application and repeated use. The reviewer would even have been forced to purchase a copy. Its positive values outweigh considerably those inclusions of dubious merit. Even the latter are of value, since they emphasize the need for more information on many common mineral species.

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FORTUNES IN MINERALS INCLUDING URANIUM. Simple Tests and How to Make Them by Ion L. Idriess. 310 pp., 3 figures, 2 plates, several tables. Anglobooks, 475 Fifth Avenue, New York 17, N. Y. $5.50, new ed., 1951.

If you like your mineralogy cookbook style, this is your dish; if you like your geology homespun, then this book is woven after your woof and warp. Mr. Idriess writes for the non-technical prospector and amateur mineralogist in a loose conversational style, seemingly intended to assure the layman that, after all, the earth sciences are simple, mundane subjects, whose principles can be learned by rote. Several introductory chapters, containing such exhortations as, Get the idea! and Use your head! are followed by general discussions of rocks and mineral deposits ("To learn the different kinds of rocks is simple—merely the memorizing of a score or so of the main rocks."). Next appear descriptions of various chemical tests—flame tests, glass tube tests, bead tests and wet tests. Two chapters are devoted to detailing the tests for detecting 25 metallic elements. Eleven of the remaining chapters of Part I describe the ores of various metals and substances including such odd bedfellows as those of Chapter XXII, The ores of mercury, antimony and mica, and of Chapter XXIII, Ores of asbestos, tantalum and niobium, manganese, zinc, chromium, iron. These descriptions are interrupted by a three-chapter interlude on how rocks and minerals were formed ("The granite will always be a granite, the diamond a diamond, etc. Although a pegmatite is a granite it will always form into a pegmatite."). The descriptive section concludes with "A summary of minerals and their families" and an afterthought chapter on the diamond. Part II deals with uranium, its ores—primary and secondary and how and where to hunt for it.

The book is full of errors of fact, of implication, of omission and of typography, as well as being garrulous, verbose and circumloquacious. It would substitute simpleness for simplicity and table d'hôte methodology for the application of scientific principles.

The last chapter, a one-page statement of the "mineral wealth" of the sea, winds up with, "And so ends this book on minerals. Read it carefully, then read it again, and the information in it will become much plainer." I doubt it.

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SIR DOUGLAS MAWSON ANNIVERSARY VOLUME, The University of Adelaide, Adelaide, Australia; 224 pp. 1952.

The Sir Douglas Mawson Anniversary Volume is a collection of 16 contributions to geology in honor of Professor Mawson's 70th birthday anniversary, presented by colleagues, friends and pupils and edited by M. F. Glaessner and E. A. Rudd. A fitting laudatory preface, provided by Emeritus Professor Sir William Mitchell, Chancellor of the University of Adelaide from 1942 to 1948, characterizes the world renowned geologist and Antarctic explorer as a resolute man and an inspiring leader.

Contributors to the volume include A. R. Alderman, D. R. Bowes, W. R. Browne, W. H. Bryan, W. B. Dallwitz, M. F. Glaessner, E. S. Hills, A. W. Kleeman, C. E. Marshall, R. T. Prider, R. C. Sprigg, F. L. Stillwell, C. E. Tilley, L. K. Ward, A. W. G. Whittle and A. F. Wilson. The following papers will be of particular interest to mineralogists and petrologists: The transformation of quartzite by migmatization at Mount Fitton, South Australia by Bowes; A note on glauconitic minerals of low refractive index from Lower Tertiary beds in South Australia and Victoria by Dallwitz; The determination of the extinction angle in monoclinic pyroxenes and amphiboles by Kleeman; Uraninite from Rum Jungle and Ferguson River, Northern Territory by Stillwell; Nepheline parageneses by Tilley; and The charnockite problem in Australia by Wilson. Other papers embrace topics in the fields of areal and economic geology, glaciation, metamorphism of coal, sedimentation, stratigraphy and tectonics. The general high quality of the collected works can be regarded as a measure of the esteem in which their inspirer is held.

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THE THIN-SECTION MINERALOGY OF CERAMIC MATERIALS, by G. R. Rigby. 179 pp., 11 figures, 20 tables, 3 indexes. Published by The British Refractories Research Association, Beechfield, Queens Road, Tenkhull, Stoke-on-Trent, Staffordshire, England. 1948, 21 s. ($2.95).

This is in several ways a most useful volume, summarizing as it does widely scattered data on crystalline compounds ("artificial minerals") and minerals encountered in the microscopic examination of ceramic raw materials, products and wastes. Although the book is designed primarily to introduce microscopic mineralogical techniques to the technical personnel of the ceramic industry, it contains much information of value to workers in mineralogy, too. The book is divided into five parts: I. The preparation of thin sections; II. The identification of mineral phases; III. Optical properties of minerals found in ceramic materials, slags, glasses and sinters; IV. Determinative tables; and V. Appendices. Part I, on thin-section preparation, touches on impregnation, cutting, grinding, mounting and covering. Ignored are some of the recent advances in thin-section technology, such as the use of methyl metacrylate (Bell, *Econ. Geol.*, **34,** 804–811, 1939) and new improved cutting methods (Meyer, *Econ. Geol.*, **41,** 166–172, 1946).

Part II is a condensed, simplified treatment of methods in crystal optics which omits the usual background in the physics of light for vectorial crystalline materials. Dr. Rigby describes the microscope and its adjustment, determination of crystal (optical) group, refractive indices and birefringence, crystal systems and habit, and optical orientation. Under refractive index determination, the immersion method of mineral study is described briefly, despite the restriction of the title of the book to "thin-section mineralogy."

Mineralogists probably will find most valuable Part III, which describes the minerals and "artificial minerals" of ceramic raw materials, slags, glasses and sinters. There are approximately 140 entries, not all of equal status, for whereas most are species, some entries are for series and a few even represent groups. The most complete descriptions include: composition, specific gravity, crystal system, optical groups and sign, melting point, refractive indices, interference figure, relief, birefringence, form, cleavage, color, distinguishing features and occurrence. The determinative tables of Part IV are separated first for the optical groups and then constructed on the basis of increasing average index of refraction. However, it is not made clear what this index value represents, whether it is a mean index for biaxial minerals or an average mean index for those minerals with considerable optical variation. Thus the tables are useful only in a very general way. Appendix

I is White's test for uncombined lime; II is the bibliography; and III is a list of references to photographs that illustrate many of the described substances.

The book has two successes: It is a praiseworthy attempt to acquaint workers in ceramic materials with a highly useful technique developed in a neighboring, but to them, often largely alien science; and also, it makes available to mineralogists information in collected form on artificial substances that supplements data on their natural counterparts.

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INTERNATIONAL TABLES FOR X-RAY CRYSTALLOGRAPHY. Vol. I. Symmetry Groups. Editors: N. F. M. Henry and K. Lonsdale. xii+558 pages, 237 figures. Published for the International Union of Crystallography. The Kynoch Press, Birmingham, England. 1952. Price £5 5s, postpaid.

The first volume of what was originally planned as a revised edition of the *International Tables for the Determination of Crystal Structures* has now appeared. Because of the many changes which have been made, it is considered to be an entirely new publication, and has a new title. The most obvious change is that the entire text is in English. However, in examining the new volume, many other significant changes are to be found.

The volume opens with a Historical Introduction by M. von Laue, which includes an account by Sir Lawrence Bragg of the early work done by himself and his father. Some of the new features which follow the introduction are as follows; sections on two-dimensional lattices, point groups and space groups, which should be of value from the pedagogical standpoint; an enlarged treatment of unit cell transformations and transformation matrices; symmetry diagrams for the 32 point groups; a sub- and super-group tabulation, the former replacing the old "Untergruppen" in each space group description; etch-figure symmetry and Laue-projection symmetry, described by the appropriate two-dimensional point group symbol, for various crystal planes in each of the 32 point groups; point group symmetry and the physical properties of crystals, including optics, optical activity, pyroelectricity and piezoelectricity.

The major new feature (158 pages) consists of the general geometrical structure factors, which in the previous edition were included in the individual space group descriptions. Also given are simplified structure factors for certain special positions of the space groups, and the electron density formulae. This section is essentially in the form previously published by Dr. Lonsdale in her *Structure Factor Tables*. In that publication, all space groups having a center of symmetry, but with the origin elsewhere, had alternative descriptions with the origin moved to the center of symmetry. Not only has this been done in this new section, but likewise has been done in the main section, where dual descriptions are given for such space groups.

In the descriptions of the 230 space groups, several changes are evident. Two descriptions are given for each monoclinic group. One (1st setting) has the unique axis in the z position, and one (2nd setting) has the unique axis in the conventional crystallographic y position. In the tetragonal system the alternative C and F descriptions are omitted, and all drawings are in the normal position; likewise with the H descriptions in the hexagonal system. Also, the hexagonal primitive lattice is very properly designated as P rather than C. In the cubic system the diagrams intended to show the general positions and the distribution of symmetry elements have been entirely eliminated.

Minor changes include a new order for some of the 32 point groups. For example; monoclinic- 2, m, $2/m$; orthorhombic- 222, $mm2$, mmm; tetragonal- 4, $\bar{4}$, $4/m$, 422, $4mm$, $\bar{4}2m$, $4/mmm$. Some of the abbreviated space group symbols have been dropped, and more complete symbols used instead, such as $mm2$, 422, $\bar{4}_2cm$, $4_2/m\ cm$, 622, 6_3cm and 432.

A tabulation of the diffraction symbols for the 230 space groups, following that of Buerger (*X-Ray Crystallography*), is included. The order of listing differs from that of Schoenflies, and somewhat from that of Buerger. Following the space group descriptions, the structure factor tables and electron density formulae, are several short sections, including Patterson and Patterson-Harker Functions; Transformation of Coordinates; and Notes on Special Topics. These topics are: Reduction of General Primitive Lattice Triplet to the Corresponding Conventional Bravais-lattice Triplet; Sub- and Super-groups of the Space Groups; Space Group Determinations by Methods Outside the Friedel Law; and Inequalities Arising from Symmetry Elements. The volume concludes with an Index of Three-Dimensional Space-group Symbols for Various Settings, and a Dictionary of Crystallographic Terms in English, French, German, Russian and Spanish.

This new volume in no way impairs the continued usefulness of the first edition. However, this improved and enlarged volume is a real contribution to this important field of science, and the editors are to be congratulated for the splendid work they and their co-workers have done. Incidentally, the quality of the printing is excellent.

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STRUCTURE REPORTS FOR 1949. Vol. 12. General Editor, A. J. C. Wilson; Section Editors: C. S. Barrett (Metals), J. M. Bijvoet (Inorganic compounds), J. M. Robertson (Organic). Published for the International Union of Crystallography. N. V. A. Oosthoek's Uitgevers Mij, Utrecht. viii+478 pp. (1952). Price 45.-Dutch florins, postpaid.

This is a companion to volume 11 (for 1947–8). Volumes 8, 9 and 10, which will fill the gap since the last *Strukturbericht* (vol. 7, 1939), are yet to appear. The editors have continued the policy of giving such a complete summary of the structure data that in most cases reference to the original article will not be necessary. On the other hand, non-structural data in the original paper, no matter how important, are not abstracted. In some cases the editors have added information, such as interatomic distances, when not given in the original. They may make critical comments concerning the data presented. Both of these features are very desirable.

The section on metals is arranged alphabetically. In the other two sections it is necessary to use either the formula or the subject index to locate a given substance. An author index is also included. The many workers who have occasion to use these reports will be grateful for the efforts of the editors and the abstractors who have made this volume possible.

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SULPHUR. A Study by the Secretariat of the Chemical Products Committee. Published by the Organisation for European Economic Cooperation. Paris. 1952. Columbia University Press, New York. 62 pp. Price $1.00.

A study of potential sulfur supplies from among the members of the O.E.E.C., including native sulfur, pyrite, oil refining, shale-oil, and coal. In spite of possible increased production, improved methods of recovery and economies in the use of H_2SO_4, there is a serious deficit in prospect. However, in terms of the U. S. production this deficit is very small. It could easily be met by an increased quota from the U. S., which in turn could easily be accomplished by employing even on a small scale certain economies—especially in the production of fertilizers which require little or no H_2SO_4.

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